

TECHNICAL REPORTS SERIES No. **324**

# Safe Handling of Tritium

## Review of Data and Experience



INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1991



# SAFE HANDLING OF TRITIUM

Review of Data and Experience

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## FOREWORD

The use of tritium has become increasingly important in several areas, for example in the manufacturing of tritium powered devices, radiochemicals and pharmaceuticals; in the operation of heavy water fission reactors; in the operation of laboratories handling or processing tritium; and in research and development related to fusion energy. Since tritium can be incorporated into body fluids and tissues and irradiate them, there is a need to provide practical guidance and recommendations for its safe handling.

This publication contains information on such topics as dosimetry and monitoring of tritium, protective clothing, safe practices in tritium handling laboratories and tritium compatible materials. The information has been compiled from current experience in various applications of tritium. It is anticipated that it will be valuable as resource material to all users of tritium, including those involved in fusion R&D.

The environmental behaviour of tritium and its impact on the public at large are not addressed in this publication, since these topics have been dealt with elsewhere by the International Atomic Energy Agency and other organizations.

This publication is the result of an IAEA Technical Committee Meeting on Safe Handling of Tritium held in Vienna from 13 to 17 October 1986 and a consultants meeting held in Chalk River, Ontario, in March 1987, with final compilation and review in December 1989.

The IAEA is grateful to the many experts who have contributed to this publication, either as participants at the Technical Committee or consultants meetings, or as contributors and reviewers.

### *EDITORIAL NOTE*

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# 1. INTRODUCTION

## 1.1. BACKGROUND

The technologies for the management of tritium bearing effluents and wastes have been addressed in two publications in the International Atomic Energy Agency's Technical Reports Series [1, 2]. Tritium was identified in a study by the Nuclear Energy Agency of the Organisation for Economic Co-operation and Development (OECD/NEA) as one of four volatile radionuclides arising from the nuclear fuel cycle with potential long term radiological significance [3]. The environmental behaviour of tritium was addressed in a symposium jointly organized by the IAEA and the OECD/NEA [4].

## 1.2. OBJECTIVES

The main objective of this publication is to provide practical guidance and recommendations on operational radiation protection aspects related to the safe handling of tritium. This publication will also serve as a framework for the exchange of information among Member States and for identifying further data or studies that may be required.

The recommendations in this publication should not be interpreted as standards but should be regarded as good practices which, if applied appropriately, could contribute to improved safety in the operation of tritium handling facilities.

## 1.3. SCOPE

This publication addresses primarily requirements for the safe handling of tritium in the following types of facility:

- (a) Tritium handling laboratories;
- (b) Industrial scale nuclear facilities such as heavy water reactors, tritium removal plants and fission fuel reprocessing plants;
- (c) Facilities for manufacturing commercial tritium containing devices and radiochemicals.

The requirements of nuclear fusion reactors are not addressed specifically, since there is as yet no tritium handling experience with them. However, much of the material covered in this publication is expected to be relevant to them as well.

Annex III of this publication briefly addresses problems in the comparatively small scale use of tritium at universities, medical research centres and similar establishments. However, the main subject of this publication is the handling of larger quantities of tritium.

Operational aspects include designing for tritium safety, safe handling practice, the selection of tritium compatible materials and equipment, exposure assessment, monitoring, contamination control and the design and use of personal protective equipment.

This publication does not address the technologies involved in tritium control and cleanup of effluents, tritium removal, or immobilization and disposal of tritium wastes, nor does it address the environmental behaviour of tritium.

#### 1.4. GENERAL PRINCIPLES OF TRITIUM SAFE HANDLING

The overall objectives of radiation protection are to prevent acute injurious exposures, to ensure that regulatory limits are not exceeded and to keep exposures as low as reasonably achievable (ALARA). Experience at several organizations has shown that the following organizational factors help to ensure the effectiveness of the radiation protection programme:

- (a) Management commitment to minimizing exposures;
- (b) An independent health and safety department to provide overall radiation safety programme direction and independent assessment;
- (c) A close relationship between facility designer and operator, which permits operating experience to be translated quickly into improved designs.

The elements of an occupational radiation safety programme for tritium are:

- (1) Source control
  - Source reduction,
  - Source containment by engineering controls.
- (2) Exposure control
  - Designing for maintenance,
  - Radiological safety procedures,
  - Work place monitoring,
  - Contamination control,
  - Waste management,
  - Personal protective equipment.
- (3) Exposure assessment
  - Bioassay,
  - Dosimetry.

- (4) ALARA programme
  - ALARA policy,
  - Design safety review,
  - Operational safety audit.
- (5) Radiation protection training.

The control of hazards at the source is in general the most effective. Tritium control may consist of the following elements:

- (i) System component leaktightness,
- (ii) Control of tritium permeation,
- (iii) Containment of tritiated atmospheres with multiple barriers,
- (iv) Confinement of tritiated waste,
- (v) Atmospheric cleanup,
- (vi) Ventilation control.

Tritium handling and exposure control depend to some extent on the type of facility (nuclear reactors, tritium laboratories, etc.), the chemical form of the tritium and the quantity of tritium handled. For large nuclear facilities, formal design safety reviews and operational safety audits will provide cost effective exposure reduction strategies.

## 2. RADIOLOGICAL HAZARDS AND DOSIMETRY

### 2.1. TRITIUM HAZARDS FROM OCCUPATIONAL EXPOSURES

Tritium can be present in the work place in airborne form as a gas, a vapour, or tritiated dust or aerosol; in contaminated liquids (oils, water); or on contaminated equipment and materials. Intake pathways of tritium into the human body are:

- (a) Inhalation;
- (b) Skin uptake
  - Diffusion of airborne tritium (mainly tritium oxide (HTO)<sup>1</sup>) through the intact skin,
  - Skin absorption through contact with contaminated surfaces or with contaminated liquids;
- (c) Ingestion.

The biological hazards of the different forms of tritium can differ markedly. Following intake into the body, HTO mixes uniformly within two to three hours with

---

<sup>1</sup> In this publication, tritium in the form of the oxide, HTO, DTO or T<sub>2</sub>O, is generally written as HTO.

the body water. It can diffuse into cells and, to a small extent, can be incorporated into organic compounds in the body. Tritium gas (HT)<sup>2</sup>, on the other hand, is not readily incorporated into body water or tissue. HT dissolved in body water is rapidly eliminated from the body, and therefore inhaled HT is mainly an irradiator of lung tissue (a relatively mild irradiator, owing to the very low energy of the tritium  $\beta$  particle (mean energy 5.7 keV)). On the basis of Publication 30 of the International Commission on Radiological Protection (ICRP), the radiological hazard of HTO relative to that of HT for equal concentrations in air is 25 000 to 1, for occupational exposure. See, however, Section 2.2.2.

HT can be converted to HTO through oxidation and isotopic exchange mechanisms. Because of the large difference in radiological hazard between HT and HTO, this conversion process is a major consideration in tritium safe handling.

Tritium present in particulate form, such as metal tritides, may be retained in the lungs when inhaled, with a biological elimination half-life that could be of the order of hundreds of days if the material has low solubility. The dosimetry is not well established and bioassay is difficult.

Other tritiated chemical species that may be present in certain work environments include hydrocarbons and ammonia. Other tritiated organic compounds are also encountered in the production and handling of tritiated luminous compounds and radiopharmaceuticals.

## 2.2. TRITIUM DOSIMETRY

### 2.2.1. Dosimetry

From the energy conversion factor,  $1 \text{ MeV} = 1.602 \times 10^{-13} \text{ J}$ , and the assumption of homogeneous tritium distribution in soft tissue, the absorbed dose rate  $\dot{D}$  is given by:

$$\dot{D} = 1.602 \times 10^{-13} EA(t)/M \quad (\text{Gy/s}) \quad (1)$$

or

$$\dot{D} = 1.384 \times 10^{-8} EA(t)/M \quad (\text{Gy/d}) \quad (2)$$

where

E is the average absorbed energy per decay (MeV);

A(t) is the time dependent activity (Bq);

M is the mass of material in which the energy E is absorbed (kg).

---

<sup>2</sup> In this publication, tritium in the form of hydrogen molecules, HT, DT or T<sub>2</sub>, as a gas or adsorbed on surfaces, is generally written as HT.



The committed absorbed dose  $D$  is

$$D = 1.384 \times 10^{-8} (E/M) \int_0^{\infty} A(t) dt \quad (\text{Gy}) \quad (3)$$

The dose equivalent to a tissue  $j$  is given by:

$$H_j = QND_j \quad (4)$$

where

$H_j$  is the dose equivalent (Sv);

$Q$  is the quality factor;

$N$  is other modifying factors.

The range of the tritium  $\beta$  particles (maximum energy 18.6 keV, mean energy 5.7 keV) in tissue is less than  $6 \mu\text{m}$  [5], which is considerably less than the depth of basal skin cells below the surface of the skin ( $\sim 70 \mu\text{m}$ ). The range of 90% of the  $\beta$  particles is less than  $1 \mu\text{m}$ . Tritium is therefore not considered hazardous unless it enters the body. Typically, mammalian cells are about  $10\text{--}20 \mu\text{m}$  in diameter, with the cell nucleus much smaller [5]. Since the range of the tritium  $\beta$  particle is less than a cell diameter, the distribution of tritium in cells can be important in evaluating the radiological hazard, because the distribution of damage could be non-uniform. This contrasts with irradiation from  $\gamma$  or higher energy  $\beta$  particles, where the distribution of energy deposition is essentially independent of structure and only the average energy deposited in tissue needs to be considered.

In addition to the possible distributional effects mentioned above, two other properties of tritium can influence its hazard. The first is that the decay of tritium in a molecule leaves the molecule with a  $^3\text{He}$  atom at the site of decay. Depending on the molecule, and the specific site in the molecule, the transmutation can be more damaging than the emission of a  $\beta$  particle. The probability of the entrance of a tritium atom from tritiated water and other general tritiated metabolites into specific and radiosensitive sites is of the order of  $5 \times 10^{-4}$  [6].

The other property of tritium that can influence its hazard is related to the distribution of ionizations and excitations along the track of the  $\beta$  particle. More dense interactions are usually considered to be more damaging than less dense interactions. This property is usually quantified by the use of linear energy transfer, or  $\text{LET}_{\infty}$  (usually denoted simply as LET), which is the average energy deposited per unit path length of the particle depositing the energy. The quality factor ( $Q$ ) of radiation is often related to the LET by the relationship [7]

$$Q = 0.8 + 0.16\text{LET} \quad (5)$$

where LET is in units of  $\text{keV}/\mu\text{m}$ .

TABLE I. VALUES OF LET AND Q FOR TYPICAL LOW LET RADIATIONS

Radiation	Approximate LET (keV/ $\mu$ m)	Quality factor, Q	
		Calculated from Eq. (5)	From ICRP Publication 26
50 kVp X rays (2.5 mm Be)	7.5	2.0	1
Tritium $\beta$ particles	5.5	1.7	1
150 kVp X rays (2.5 mm Be)	2.5	1.2	1
150 kVp X rays (0.3 mm Cu)	1.5	1.0	1
300 kVp X rays	1.0	0.9	1
Co-60 $\gamma$ rays	0.84	0.8	1

However, the ICRP has recently reviewed this relationship [8–10] and reiterated its previous conclusion [11] that there was no justification for using a quality factor different from one for all ‘low LET’ radiation. Experiments that measure the relative biological effectiveness (RBE) as a function of LET, dose and dose rate must be taken into account as well as the physical definition of Q given above. Values of LET and Q for some typical low LET radiations are given in Table I.

The ICRP had earlier recommended a quality factor of 1.7 for tritium [12], and this value may still be used by some countries. However, most countries have accepted the later Recommendations of the ICRP [8–11] and currently use a quality factor of 1 in dosimetry calculations for tritium.

### 2.2.2. Tritium oxide

A general approach for the calculation of dose resulting from tritium incorporation in the human body is to apply a metabolic model which is an oversimplification of what could happen in practice but which is sufficiently accurate for the derivation of intake limits for workers. In the model recommended in ICRP 30 tritiated water is assumed to be distributed uniformly and instantaneously among all soft tissues at any time following the intake, independently of the routes of entry. Its retention is described by a single exponential with a half-life of 10 d. Thus in the case of a single

intake of  $A_0$  Bq of HTO into the human body, its concentration in soft tissue (total weight 63 kg) is given by:

$$C(t) = \frac{A(t)}{63} = \frac{A_0}{63} \exp\left(-\frac{\ln 2}{10} t\right) \quad (6)$$

where  $C(t)$  and  $A(t)$  are, respectively, the concentration (Bq/kg) and the body content (Bq) at time  $t$  (d) after uptake. The committed absorbed dose  $D$  is:

$$\begin{aligned} D &= 1.384 \times 10^{-8} \text{ E} \int_0^{\infty} \frac{A_0}{63} \exp\left(-\frac{\ln 2}{10} t\right) dt \\ &= 1.807 \times 10^{-11} A_0 \text{ (Gy)} \end{aligned} \quad (7)$$

Hence:

$$ALI = \frac{H_L}{1.8 \times 10^{-11}} \quad (8)$$

where

$ALI$  is the annual limit of intake (Bq);

$H_L$  is the annual limit of effective dose equivalent (Sv).

With  $H_L = 0.05$  Sv and  $Q = 1$ ,

$$ALI = 2.78 \text{ GBq} \approx 3 \text{ GBq}$$

It was shown that exposure to an atmosphere contaminated by HTO vapour results in the permeation of tritium through the intact skin [13]. In the model recommended by the ICRP, the ratio of intake by skin absorption to intake by inhalation is assumed to be 0.5 for occupational exposures. Therefore, for the calculation of the derived air concentration (DAC), the assumed inhalation rate of  $0.02 \text{ m}^3/\text{min}$  must be multiplied by 1.5 to obtain the total tritium intake. Assuming 2000 working hours per year, one obtains:

$$\begin{aligned} DAC &= \frac{ALI}{1.5 \times 0.02 \times 60 \times 2000} \\ &= 7.69 \times 10^5 \text{ Bq/m}^3 \approx 8 \times 10^5 \text{ Bq/m}^3 \end{aligned} \quad (9)$$

Dosimetric calculations for HTO are summarized in Table II.

TABLE II. DOSIMETRY OF HTO BASED ON ICRP PUBLICATION 30

Reference tissue	Soft tissue
Mass	63 kg
Effective half-life	10 d
Effective energy	0.0057 MeV
Quality factor Q	1.0
Dose per unit intake	$1.8 \times 10^{-11}$ Sv/Bq
Ratio of skin absorption intake to inhalation intake	0.5
ALI	$3 \times 10^9$ Bq/a
DAC	$8 \times 10^5$ Bq/m <sup>3</sup>

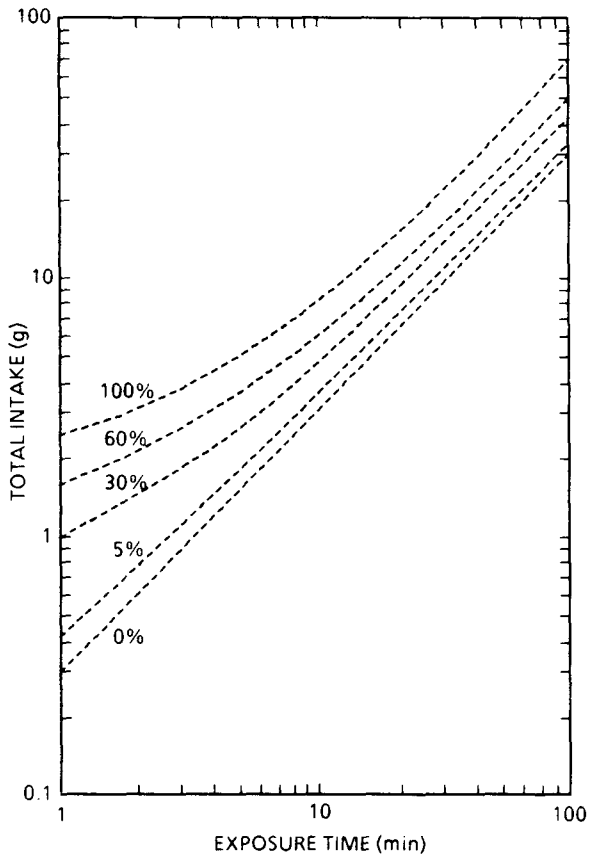


FIG. 1. Total intake of tritiated water with various percentages of skin area wetted.

When the skin is splashed with tritiated water, intake through the wetted skin by means of the blotter effect also occurs (Fig. 1). Tritiated water soaks into the extravascular layer of skin, like ink into blotting paper. Some of the soaked in HTO remains in the extravascular layer even after washing and subsequently diffuses into the body [14]. Total intake to the body is the sum of three intake pathways [15]:

$$\text{Respiratory intake} = RTHN \quad (\text{Bq}) \quad (10a)$$

$$\text{Non-wetted skin intake} = (S - W) ITHN \quad (\text{Bq}) \quad (10b)$$

$$\text{Wetted skin intake} = MWN + WIPNT \quad (\text{Bq}) \quad (10c)$$

where

- R is the respiration rate (20.8 L/min);
- T is the time of exposure (min);
- H is the humidity of the air (g/L);
- N is the specific activity of tritiated water and water vapour (Bq/g);
- S is the total skin surface area (1.9 m<sup>2</sup>);
- W is the wetted skin surface area (m<sup>2</sup>);
- I is the intake rate through skin diffusion (5.1 L·m<sup>-2</sup>·min<sup>-1</sup>);
- M is the intake due to the blotter effect (1 g/m<sup>2</sup>);
- P is the humidity of the air at skin temperature (0.04 g/L).

### 2.2.3. Tritium gas

ICRP 2 [12] considered HT to be a skin irradiator and derived a value for maximum permissible concentration in air (MPC<sub>a</sub>) of 2000 μCi/m<sup>3</sup> (7.4 × 10<sup>7</sup> Bq/m<sup>3</sup>) using the semi-infinite cloud model. This is unduly conservative since, as previously pointed out, the maximum range of the tritium β particles is about 6 μm and the depth of basal skin cells is about 70 μm.

ICRP 30 [10] considered doses received by the lung from HT contained in the lung to be the major exposure pathway. Using an average lung volume of 0.003 m<sup>3</sup> and lung mass of 1 kg, the DAC was calculated to be 2 × 10<sup>10</sup> Bq/m<sup>3</sup>. The ratio of the DAC for HT to that for HTO for occupational exposure is 2 × 10<sup>10</sup>/8 × 10<sup>5</sup> = 25 000.

ICRP 30 did not consider the dose from HTO produced from in vivo oxidation of the small amount of HT absorbed in the body. Pinson and Langham [14] reported from their experimental findings that both man and rat slowly oxidized inspired HT into HTO. Hydrogenase-bearing bacteria in the intestinal tract were considered to be the principal agent responsible for in vivo oxidation. Experimental work at the Chalk River Nuclear Laboratories (CRNL) in Canada confirmed these earlier findings [16]. It was shown [16, 17] that the effective dose from HTO produced by in vivo oxidation is approximately equal to that from lung irradiation by HT and hence

the ratio of the DAC for HT to that for HTO should probably be closer to  $10^4$ . On the other hand, the dose to the lung may be somewhat overestimated since the radio-sensitive cells in the lung may be somewhat shielded by overlying tissue.

#### 2.2.3.1. Skin contact

Another hazard in handling HT is the skin uptake of tritium from contact with surfaces contaminated with HT. Early work by Eakins et al. [18] showed that tritium entering the body through skin contact with HT contaminated metal surfaces was excreted in urine primarily in organic forms. More recent experimental work at CRNL [19] using animals confirmed elevated levels of organically bound tritium (OBT), especially in skin. During the first day following exposure the skin OBT concentration was at least 20 times higher than that in the liver, the tissue with the next highest concentration, and the ratio of OBT to HTO in skin was about 30. The dosimetry of tritium from tritium contaminated surfaces therefore reduces to estimating the dose to the skin at the point of contact.

Johnson and Dunford [20] developed models to fit the data of Eakins et al. [18] and showed that the dose to a 0.1 cm layer in the dermis, assuming an exposed area of 40 cm<sup>2</sup>, ranged from  $4.5 \times 10^{-9}$  to  $4.5 \times 10^{-8}$  Sv per becquerel uptake. In Ref. [18] the fraction of contacted HT contamination which was taken into the body was found to range from 0.004 to 0.014 in four subjects whose forearms were exposed. In two subjects whose palms were exposed, the fractional uptakes were 0.012 and 0.0072.

On the basis of an average uptake fraction of 0.01 and a skin dose factor of  $4.5 \times 10^{-8}$  Sv per becquerel uptake, an HT surface contamination limit of  $1.5 \times 10^7$  Bq/m<sup>2</sup> (500  $\mu$ Ci/m<sup>2</sup>) has been derived for use at Ontario Hydro, in Canada. This limit corresponds to the ICRP dose limit of 0.5 Sv per year for any single organ or tissue [8–10]. It is based on the conservative assumption that a worker will contact the contaminated surface 10 times per day in each of the 250 working days per year. It is worth noting that the ICRP, in recommending surface contamination limits for the handling of radioisotopes in hospitals and medical research establishments [21], provides a limit of 10  $\mu$ Ci/m<sup>2</sup> ( $3.7 \times 10^5$  Bq/m<sup>2</sup>) for general radionuclide contamination on working surfaces, but states that this limit reasonably can be increased by a factor of 100 (to 1000  $\mu$ Ci/m<sup>2</sup> ( $3.7 \times 10^7$  Bq/m<sup>2</sup>)) for tritium and <sup>14</sup>C.

#### 2.2.4. Metal tritides

HT is commonly immobilized and stored in the form of metal tritides. Typical metals used are uranium, titanium and zirconium. When suspended as fine particulates in air, metal tritides could present inhalation hazards. These were not considered by the ICRP. The problem is dealt with in Refs [22, 23].

A preliminary assessment of the radiological hazard from metal tritide particulates was made by Ontario Hydro. Two potential hazards were considered:

- (a) Direct irradiation of lung cells by  $\beta$  particles emitted from metal tritides deposited in the lung;
- (b) Leaching of tritium from the particles into body fluid, followed by oxidation to HTO.

The hazard decreases with increasing particle size, which results in increased self-absorption of the tritium  $\beta$  within the tritide particles and a decreased leach rate of tritium into body fluids. The retention half-life of metal tritide in the lung is uncertain; it would be about 500 days if Class Y compounds are assumed. On the basis of this model and an assumed mean particle size of 1  $\mu\text{m}$ , the effective dose per becquerel intake of metal tritide was calculated to be about 40 times higher than that for HTO. The calculation was conservative, owing to the fact that the shielding provided by intervening lung structures between the metal tritides and radiosensitive lung cells was disregarded. De Ras et al. [22] tentatively concluded that for inhaled titanium tritide particles the dose is considerably smaller than in cases where tritium is incorporated in comparable quantities of HTO. An acceptable dosimetric model for metal tritides is not yet available.

### 2.3. BIOASSAY PRACTICE

The concentration of HTO in the body is determined by analysing the HTO concentration in urine, a procedure commonly referred to as 'bioassay'. The dose rate to all organs is proportional to the concentration of HTO in urine. In a single intake, the committed dose equivalent (H) is obtained from Eq. (3) by setting  $E = 0.0057 \text{ MeV}$ :

$$\begin{aligned}
 H &= 7.890 \times 10^{-11} (1/M) \int_0^{\infty} A_0 e^{-\lambda_e t} dt \\
 &= 7.890 \times 10^{-11} \frac{A_0}{M\lambda_e}
 \end{aligned} \tag{11}$$

where

- $A_0$  is the total activity in the body at time zero,  $t = 0$  (Bq);
- $\lambda_e$  is the effective elimination constant ( $\text{d}^{-1}$ );
- $t$  is the time after intake (d).

$$\lambda_e = (\ln 2)/T_{\text{eff}} \tag{12}$$

where  $T_{\text{eff}}$  is the effective half-life (d).

$$\frac{1}{T_{\text{eff}}} = \frac{1}{T_r} + \frac{1}{T_b} \approx \frac{1}{T_b} \quad (13)$$

where

$T_r$  is the radiological decay half-life (12.3 a);

$T_b$  is the biological half-life (d).

$T_b$  may be obtained from bioassay measurements. If  $T_b$  is not obtained by measurement, then the average value of 10 d could be used.

A more precise evaluation model was developed by Johnson [24]. The effective dose equivalent rate ( $\dot{H}$ ) and the committed effective dose equivalent as a function of time  $t$  ( $H$ ) are given by the following equations:

$$\dot{H}(t) = 5.8 \times 10^{-11} C_u(t) \quad (\text{Sv/d}) \quad (14)$$

$$\begin{aligned} H(t) &= 5.8 \times 10^{-11} (10/\ln 2) C_u(t) \\ &= 8.4 \times 10^{-10} C_u(t) \quad (\text{Sv}) \end{aligned} \quad (15)$$

where  $C_u(t)$  is the concentration of HTO in urine at time  $t$  (Bq/L).

In most practical situations HTO intake will not be at a constant rate but will consist of an irregular sequence of individual intakes. Measurements should be made often enough that doses can be estimated with sufficient accuracy. Sometimes more frequent measurements may be needed for dose control purposes.

The sampling frequency of a routine bioassay programme is related to the effective half-life of HTO in the body (about 10 d) as well as the likelihood of tritium intakes. For persons working under Working Condition A as described in IAEA Safety Series No. 9, 1982 Edition [25], the following guide is suggested [26, 27]:

<i>Sampling frequency</i>	<i>Work group</i>
Weekly	Personnel who regularly perform work with tritium
Monthly	Personnel based in the radiation area but who normally do not perform work with tritium
Quarterly	Other staff at the facility

Reference [28] showed that the accuracy requirements for HTO dose calculations can be met by using the linear interpolation approximation if urine samples are obtained twice a month. In this method successive points in a urine tritium concentration (Bq/L) versus time (d) plot are joined by straight lines and the area under the joined lines (in Bq·d/L) is calculated. The committed effective dose equivalent ( $H$ ) is given by:

$$H = 5.8 \times 10^{-11} \times \text{area under joined lines} \quad (\text{Sv}) \quad (16)$$



Additional urine samples should be submitted for bioassay following known or suspected intakes. Samples should be submitted 1–2 h following intake, soon after an initial voiding, so that tritium in the bladder will be representative of the concentration of tritium in body fluids.

A bioassay quality assurance (QA) programme should be implemented as an integral part of the bioassay programme. In addition to routine laboratory quality control procedures, a regular cross-comparison programme with an accredited QA agency should be in place.

In many facilities practical bioassay control levels have been established for work restriction and for removal of employees from tritium work. For example, at Ontario Hydro the removal level is  $1.8 \times 10^6$  Bq/L (50  $\mu$ Ci/L).

Exposures to HT result in a dose to the lungs from HT in the air in the lung, as well as a dose from the HT that is converted to HTO inside the body. An upper limit of the lung dose ( $H_L$ ) from HT may be estimated from urine analysis by assuming all of the HTO measured in urine to have resulted from the in vivo conversion of inhaled HT to HTO. Experiments with human subjects [16] showed an average ratio of HTO in urine to integrated HT exposure of  $2.4 \times 10^{-8}$  Bq·L<sup>-1</sup> per Bq·m<sup>-3</sup>·min. For exposures which are short compared with the effective half-life of HTO in the body,

$$H_L = 0.8 \times 10^{-8} C_u \quad (\text{Sv}) \quad (17)$$

where  $C_u$  is the concentration of HTO in urine (Bq/L).

This lung dose may be multiplied by the weighting factor of 0.12 for lung [8–10] to give an effective dose equivalent of  $9.6 \times 10^{-10} C_u$  Sv. This value of effective dose due to lung irradiation by inhaled HT is approximately the same as the effective dose equivalent from the intake of HTO given in Eq. (15). The total effective dose equivalent is the sum of the effective dose equivalents from lung irradiation and from the HTO in the body.

The upper limit of the lung dose given above is overly conservative in most exposure situations where some (or all) of the HTO in urine may have resulted from exposure to HTO rather than HT. Direct monitoring of the airborne concentrations of HTO and HT in the work place will be useful in improving the dose estimate.

For skin contact with HT contaminated surfaces the tissue that receives the highest dose is skin (see Section 2.2.3.1). The following relationship between skin dose and urinary excretion of OBT may be derived from animal data [19] and has been proposed as an interim Ontario Hydro standard for use in interpreting bioassay data:

$$H_s = 1 \times 10^{-7} C_{\text{OBT}} \quad (\text{Sv}) \quad (18)$$

where

$H_s$  is the skin dose (Sv) averaged over  $100 \text{ cm}^2$  of skin;  
 $C_{\text{OBT}}$  is the concentration of OBT in urine (Bq/L) obtained 1 d after skin contact exposure.

There are questions as to the extent to which animal (rat) data and the relationship in Eq. (18) are applicable to humans. Johnson et al. [19] reported that about 80% of the total OBT that will be excreted by rats following exposure to HT contaminated surfaces is excreted in the first 24 h, and Eq. (18) was based on this observation. However, analysis of the data of Eakins et al. [18] on humans suggests that between 30 and 40% of the total OBT is excreted in the first 24 h. These data [18] also indicate that decontamination of the skin starting about 30 min after contact alters the OBT excretion pattern such that the maximum OBT excretion rate occurs at about 0.3–0.4 d rather than at 1 d, that the fraction of OBT excreted in the first 24 h is also decreased, and that the concentration of urinary OBT determined from a spot sample after 24 h would be less than the concentration determined from total urine collected during the first 24 h. The use of Eq. (18) under these circumstances would underestimate the dose by a factor of 3 or more.

Equation (18) also assumes that the area of contaminated skin is  $100 \text{ cm}^2$ . This assumption may result in an underestimation of the skin dose in some cases, and it would be more prudent to assume a surface area of about  $30 \text{ cm}^2$  (about 25% of the surface area of the palm and fingers of one hand). It should be noted that the measurement of the contaminated area of the skin is very uncertain (using ‘swipes’) and may result in under- or overestimation of skin doses by a factor of 10 or more. Bioassay data do not provide information on the surface area to be used when calculating skin doses.

Because of the uncertainties discussed above, it would be prudent to use the following relationship between urinary OBT concentration and skin dose rather than Eq. (18):

$$H_s = 1 \times 10^{-6} C_{\text{OBT}} \text{ (Sv)} \quad (18a)$$

where  $H_s$  and  $C_{\text{OBT}}$  are as defined for Eq. (18). The surface area is assumed to be  $30 \text{ cm}^2$ .

The distribution of OBT in time and with skin depth also requires further investigation.

## 2.4. DOSE COMMITMENT REDUCTION

### 2.4.1. Washing after exposure

Measurements at CRNL [29] showed that following acute exposure to tritiated water vapour some of the HTO can be washed out of the skin if washing is started

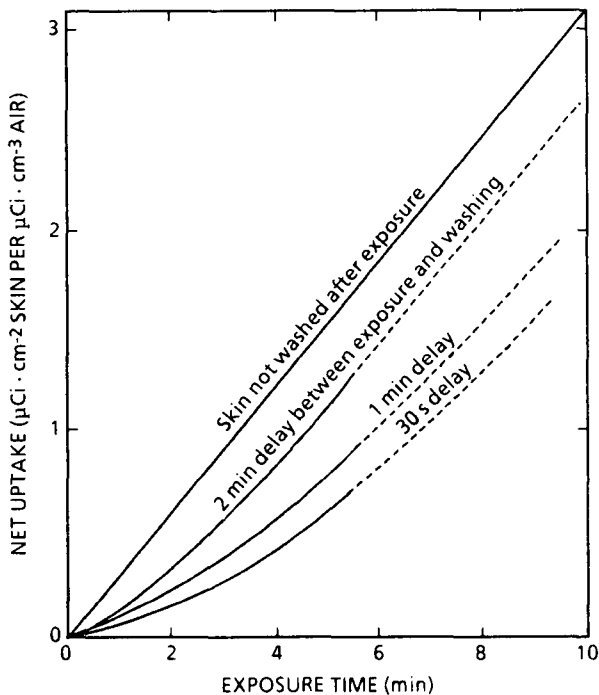


FIG. 2. Reduction of HTO uptake by washing after exposure to HTO vapour.

soon after exposure, before the HTO can reach the bloodstream and be distributed throughout the body. Volunteers were exposed to HTO vapour for periods of 30 s to 5 min, then after a delay of 30 s to 2 min the skin was sprayed with clean water for 2 min. The effect of delay between exposure and washing is shown in Fig. 2.

#### 2.4.2. Increased water intake

The committed dose from HTO already in the body can be reduced by increasing the individual's water intake rate, which in effect reduces the effective half-life of tritium in the body. Figure 3 shows the calculated dose reductions for various durations of increased water intake. The normal intake rate has been assumed to be 1.95 L/d. It can be seen that a dose reduction of up to 50% can be realistically achieved and that the marginal benefit beyond a treatment period of 10 d is minimal.

In a manner similar to the reduction in committed dose after the skin is wetted with HTO, skin doses may be reduced by washing the skin shortly after contact with

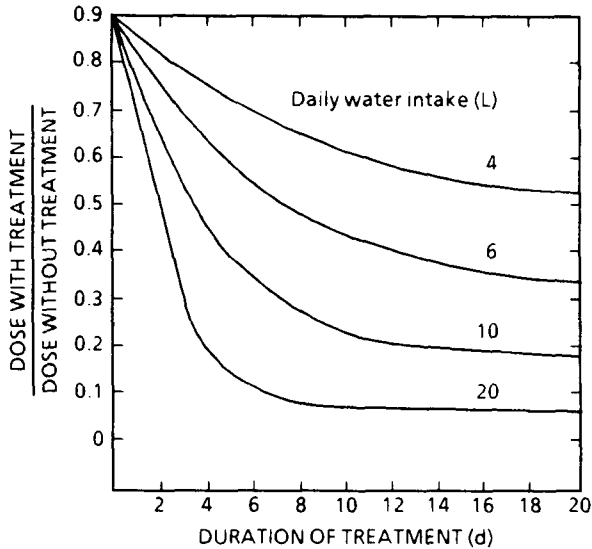


FIG. 3. Tritium dose reduction from increased water intake. Treatment started 0.5 d after acute uptake.

HT contaminated surfaces. Eakins et al. [18] observed that washing the contaminated area of the skin of one human volunteer 30 min after contact with an HT contaminated brass surface reduced the total quantity of OBT excreted in urine by a factor of about 9, while Johnson et al. [19] reported that decontamination of rat skin 1 min after contact reduced the total quantity of tritium taken up to the skin by at least one order of magnitude. These two observations taken together suggest that decontamination of the skin within about 30 min will reduce the committed skin dose by about a factor of 10 or more.

#### 2.4.2.1. Guidelines

Interim guidelines have been developed by the Health and Safety Division of Ontario Hydro for the administration of a programme of dose reduction through increased water intake. These are summarized below.

- (a) A tritium dose reduction programme is entered voluntarily.
- (b) An acute uptake that could give a committed dose of 20 mSv or more (on the basis of a 10 d half-life) is reason for initiating the programme.
- (c) Fluid intake is increased to about 4 L in the first day as comfort allows. A staff physician is called within 24 h for further guidance on the duration and intensity of the treatment. A daily fluid intake of 6–8 L is a reasonable target.

- (d) Medical assessment should be carried out as a part of the programme. This consists of:
- An initial assessment within the first 24 h, including a history of past medical problems (heart disease, hypertension, kidney disease, diabetes mellitus, bladder or prostate problems); urine dip stick assessment of renal function (sugar and protein); assessment of blood electrolytes (potassium, sodium, chloride and bicarbonates), as well as BUN (blood urea nitrogen) and creatinine;
  - Daily weight records to check for fluid retention;
  - A daily health check by a nurse to detect any adverse effects of treatment;
  - A daily urine tritium analysis to determine biological half-life and dose;
  - A physical examination and repeat blood work as directed by the physician.

#### 2.4.2.2. Experience

Two well documented cases of tritium dose reduction by increased water intake at Ontario Hydro [27] are reviewed here.

##### *Case 1*

The subject was splashed with moderator system heavy water containing about 800 GBq/kg (21.6 Ci/kg) tritium following equipment failure. His left arm, shoulder and side were wetted. The time from first wetting to showering was about 5 min. Bioassay samples submitted following the incident showed an initial urine tritium concentration of 16 MBq/L (440  $\mu$ Ci/L), which would result in a committed dose of 18 mSv, assuming a 10 d half-life.

During the first 10 d following the uptake, the effective half-life was reduced to about 4 d through a controlled programme of increased fluid intake. The programme was begun following an initial medical assessment which showed no history of adverse health problems. Detailed daily records of fluid intake, excretion and body weight were kept. Daily fluid intake was increased from 4 L on day 1 to 6 L on day 2 and remained at that level for the rest of the regimen. The subject did not think he would be able to manage more. His body weight remained constant and his renal function remained normal. There was no change in his electrolyte levels and no evidence of fluid retention. Other than some bloating and early adaptation to forcing himself to drink, the subject noticed no particular side effects.

It can be estimated from Fig. 3 that a dose reduction of about 40% was achieved as a result of the treatment.

## Case 2

The subject was sprayed with tritiated heavy water during a spill following equipment failure. His initial urine tritium level was 9 MBq/L (240  $\mu$ Ci/L).

The dose reduction programme was started approximately 2 d after the acute uptake, following medical assessments which indicated no impediments to the initiation of the programme. Fluid intake was increased by 2 L on the first day and by 4 L on the second day to a total intake of 6 L/d. The programme was maintained for 12 d, during which the subject's effective tritium half-life was reduced from 13 d to about 5.5 d. On the basis of daily bioassay sample results, the reduction in dose during the treatment period was calculated to be 34% and the reduction in total committed dose 19%.

The subject's body weight remained constant and there were no medical problems during the treatment period.

### 2.4.3. Other medical procedures

Besides oral intake, another means of achieving forced diuresis is by intravenous infusion of large volumes of electrolyte solutions (together with administration of furosemide); obviously, this also requires careful monitoring of the patient's fluid and electrolyte balance. Moreover, tritium excretion can be accelerated substantially by more invasive medical treatment. Such treatment should be reserved for an experienced physician and should be instituted only after careful consideration of the estimated tritium exposure, the attainable dose commitment reduction, the medical risks involved and specific contra-indications which the patient may have.

In haemodialysis [30], diffusion across a semipermeable membrane and ultrafiltration by hydrostatic pressure can be combined to remove large amounts of water and other low molecular weight substances. Using this method, biological half-lives of 4 h have been achieved [30]. If haemodialysis devices are not available, peritoneal dialysis may be an alternative, with resulting half-lives of about 13 h [30]. While haemodialysis/haemofiltration is effective in removing water and dialysable compounds, other methods are known in medicine to eliminate high molecular weight or plasma-protein-bound substances from circulating blood. These methods include haemoperfusion through activated charcoal or exchange resin, plasma exchange treatment using cell separators and exchange transfusion.

One reported case of the application of forced diuresis involved the accidental inhalation of tritiated water by two people working in the tritium light source manufacturing industry [31]. The two subjects respectively inhaled about 35 GBq and 1 GBq of tritiated water. Committed dose equivalents of about 0.8 Sv and 0.025 Sv were to be expected without any countermeasures.

Forced diuresis was commenced 100 h after the accident with a venous infusion of about 7 L/d. Diuresis was enhanced by giving 40 mg of furosemide intravenously each day. Daily measurements of routine blood parameters, electrolytes and renal functions were made and all were found to remain within normal limits. Forced diuresis was stopped after 4 d, over which period it was calculated that about 15 GBq of tritium had been excreted in urine in the high uptake case. The committed effective dose equivalent based on urine bioassay data was found to be 0.47 Sv, which represents a dose reduction of more than 40%. Medical surveillance was continued for 178 d after the incident.

### **3. TRITIUM MONITORING**

#### **3.1. INTRODUCTION**

Instrumentation and techniques for monitoring tritium have been previously reviewed [32, 33], and a detailed discussion of these is not necessary. This section addresses the principles and practical aspects of tritium monitoring in the work place and describes the common types of monitors available.

Most of the monitoring requirements can be subdivided into the monitoring of tritium in air and gases, in liquids and on surfaces. Each of these will be discussed separately with emphasis on the more common methods and the various interferences that can occur. The discrimination between HT and HTO will also be addressed.

Process monitors used to provide process data and control will not be discussed as they are generally not directly relevant to radiological protection monitoring. Also, monitors and samplers for measuring environmental tritium will not be discussed.

#### **3.2. GENERAL PRINCIPLES OF MONITORING**

To protect the worker and the environment, large quantities of tritium are usually doubly and sometimes triply contained. In the primary containment, process monitors and other instrumentation provide information for process control. The secondary containment — glove boxes, interspaces between double-wall pipes, etc. — should be monitored for any increase in tritium concentration that may indicate abnormal situations that require attention and correction. The tertiary containment is usually the working environment and is the area which normally requires monitoring for radiological protection.

The overall objective of work place monitoring is to provide exposure control and protection for the workers. Routine monitoring during normal or continuous

operations provides data confirming that the work place is safe and gives a prompt indication of changes in radiological conditions. In special operations or maintenance activities, monitoring provides a prompt indication and warning of sudden unexpected increases in tritium concentrations. Monitoring also provides data for planning work from a radiological perspective, for example determining the planned duration of stay in areas with high concentrations of tritium, or assessing the type of personal protective equipment required for a particular job.

It is common practice to locate monitors at strategic points close to the sources of tritium. In a large room which contains several tritium processing systems or banks of glove boxes, a cost effective arrangement is to connect a series of sampling points through a manifold system to one of several area tritium monitors. This provides prompt warning of changes in tritium levels over broad areas in the room. To identify the source of a suspected release, valving of the individual sampling lines is required. Alternatively, the source may be located with a portable tritium monitor.

Surface contamination in both secondary and tertiary containments can be a potential tritium uptake pathway for personnel. Direct monitoring of surfaces is feasible under well defined conditions but it is more usual to take surface smears and count these with liquid scintillation counters or gas proportional counters.

If tritium contaminated materials are abraded or machined, tritiated dust particles may result. The handling of powders or tritiated compounds may also give rise to airborne contamination. For these cases, air samples should be collected in the work environment and counted in a similar way to smears.

The atmosphere in the secondary and tertiary containments will often be eventually released to the environment, preferably through a stack to provide a wide dispersal of the tritium. Stack monitors are usually required to measure the total release in a given time, to demonstrate compliance with regulatory release limits (compliance monitoring) and to provide prompt warning of a sudden release that may exceed regulatory or control limits and may require action in the facility and outside it (control monitoring).

Compliance monitoring generally requires high sensitivity and accuracy, whereas control monitoring requires fast response time. It may not be practicable to have one instrument meeting all of these requirements. Compromises between sensitivity/accuracy and response time may have to be made. Alternatively, separate devices may be used. Compliance monitoring may involve simple, inexpensive field sampling followed by laboratory analysis, whereas control monitoring usually consists of on-line continuous readout instrumentation.

Although on-line tritium monitors for liquid effluents have been developed, it is more common to take representative samples of the liquid effluent and measure these in a liquid scintillation counter.

In large tritium handling facilities there may be large amounts of on-line tritium monitoring data. It is useful to combine these data in a computer and provide real time readout in a centralized location. This information can be particularly use-



ful in an emergency, when the source of the release can be promptly identified and corrective actions taken.

### 3.3. MONITORING OF TRITIUM IN AIR OR GAS

#### 3.3.1. Bubblers and passive samplers

To sample low levels of tritium, bubblers are commonly used. For HTO, a sample of the air to be measured is bubbled through a gas washing bottle containing distilled water or ethylene glycol. Most of the water vapour in the sample exchanges isotopically with the water in the bubbler. After a sufficient interval, the total tritium content of the water is determined by liquid scintillation counting. From the flow rate and the collection time, the total volume of air from which the tritium was washed is known. This allows the average tritium-in-air concentration to be calculated.

To measure HT, the air is generally routed through a heated precious metal catalyst (e.g. palladium) to oxidize the HT to HTO before it reaches the bubblers. By employing two or three bubblers to remove and measure the HTO, followed by a catalyst and additional bubblers for HT, both HT and HTO can be measured in the same sample of air.

A simple bubbler system consists of a filter, bubblers in series, a flow meter and a pump with flow control. The flow meter must be calibrated while the pump is operating in order to avoid possible errors due to pump pulsations. For a single 250 mL gas washing bottle filled with 200 mL of distilled water and with an air flow rate of a few litres per minute, collection efficiencies well above 90% are typical

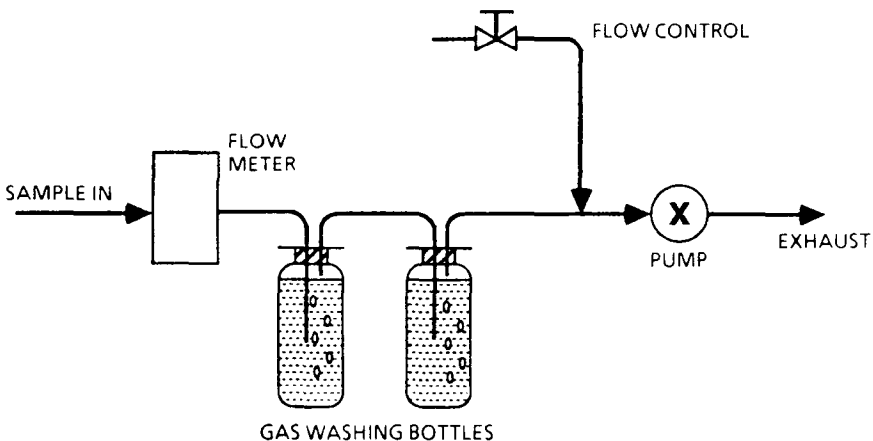


FIG. 4. Tritium sampling bubblers (flow rate 1-2 L/min). A filter may be added.

[34]. This usually precludes the necessity of making collection efficiency corrections, especially when two bubblers in series are used (Fig. 4).

Bubblers are well suited to measuring low concentrations of tritium. Even when no great care is exercised, it is easy, using liquid scintillation counting, to measure tritium in (clean) water at concentrations of about 1 Bq/mL (a few tens of picocuries per millilitre). If it is assumed that the collection volume is diluted to 1 L prior to measuring, such a concentration would result from a two hour collection time, with a flow rate of 3 L/min and a tritium-in-air concentration of about 5 kBq (100 nCi) per cubic metre. Much lower concentrations can be measured by using longer collection times or higher flow rates. For example, extending the collection time to one day decreases the detection limit by an order of magnitude.

Other radioactive gases also have some limited solubility in water. The concentration of these gases in the collected sample can be greatly reduced by purging the collected water with clean air prior to measuring. Of course,  $\gamma$  fields at the collection location will have no effect whatsoever on this method of tritium determination.

For long collection times, exceeding a few days, loss of water in the bubblers due to evaporation can become a problem. In this case the bubblers may be filled with a low vapour pressure solvent such as ethylene glycol. However, even with ethylene glycol, tritium losses have to be considered when very long collection times are used. Such losses can be reduced by having several bubblers in series and counting the tritium in each bubbler. For an HTO-catalyst-HT bubbler system, the glycol bubblers can dry the sampled air and cause the HTO formed from the oxidation of HT to adsorb onto the plumbing between the catalyst and the first HT bubbler, resulting in a delayed response to the sampled HT. One solution is to moisten the air by using water instead of glycol in the last HTO bubbler before the catalyst, but this partly defeats the purpose of using glycol instead of water.

When ethylene glycol is used as the collection liquid, the HTO is trapped by two separate processes. Firstly the HTO dissolves in the ethylene glycol and secondly there is an isotopic exchange. Some liquid scintillants will tolerate a higher loading by an ethylene glycol-water mixture than by water alone. This can lead to higher sensitivities [35]. Some cocktails are not miscible with glycol and can lead to counting difficulties. Ethylene glycol also allows the operation of bubblers at temperatures below freezing.

Diffusion samplers are beginning to be used more widely owing to their simplicity. Liquid scintillators, silica gels, zeolites and ethylene glycol have all been suggested [36, 37] for use in the sampling of HTO from air. A very simple diffusion sampler consists of an open beaker of distilled water. For short exposure times the concentration of HTO in the beaker is proportional to the HTO concentration in air and to the exposure time.

In one application of this principle the sorbent can be contained within an HTO permeable flexible plastic envelope. The sorbent could be distilled water, for example. An example of such a sampler is shown in Fig. 5.

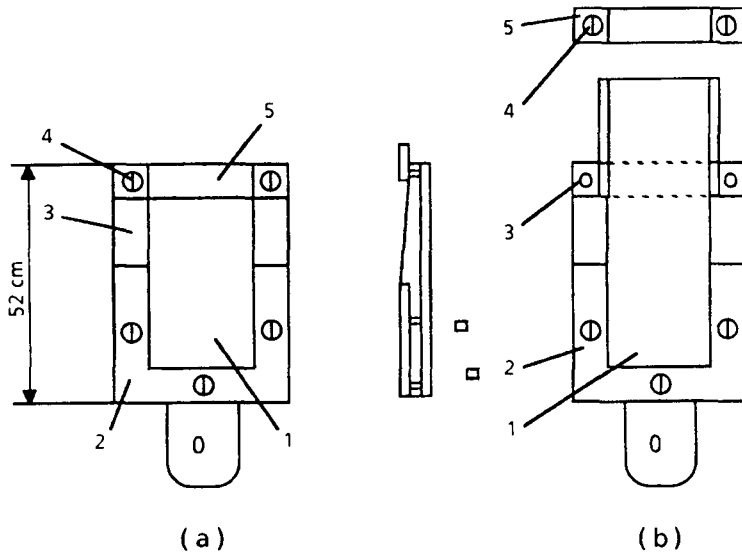


FIG. 5. Schematic drawing of diffusion sampler with distilled water in envelope in assembled form (a) and opened form (b). 1: PVC film packer; 2: strap plate; 3: housing; 4: M2 screws; 5: seating strip.

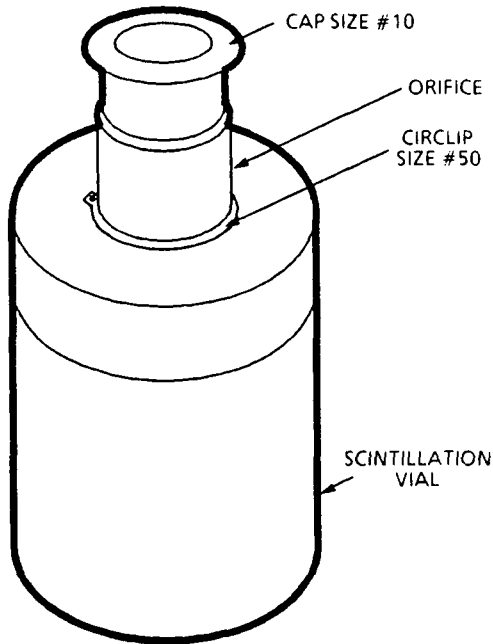
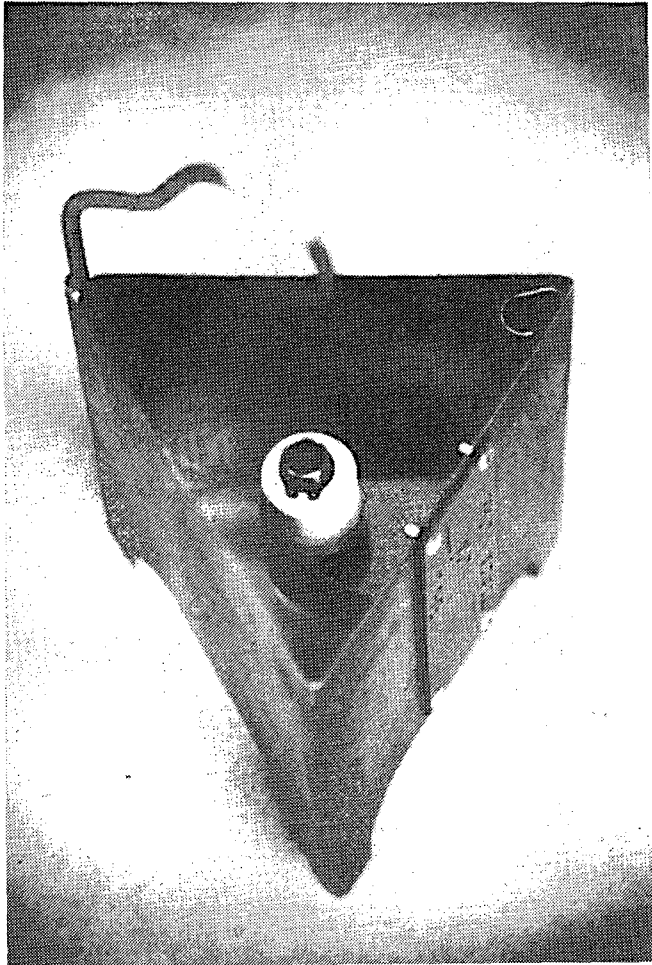


FIG. 6. Passive diffusion sampler for HTO vapour.



*FIG. 7. Passive sampler for HTO in air.*

Another application of this principle is the passive sampler for HTO in air developed by Ontario Hydro [38]. The device contains a small quantity of water, which, through exchange, will pick up HTO from the air. No pump is necessary. The exchange rate is diffusion limited and reproducible. The sensitivity of the sampler is about 15 counts/min per  $\text{kBq} \cdot \text{h}/\text{m}^3$  (500 counts/min per  $\mu\text{Ci} \cdot \text{h}/\text{m}^3$ ), with an overall accuracy of  $\pm 30\%$ . This sampler has largely supplanted bubblers at Ontario Hydro's nuclear generating stations (Figs 6, 7).

Solid desiccant (e.g. silica gel) columns are also used to collect HTO-in-air samples [39, 40]. The HTO is driven from the desiccant by heating and is then collected in a cold trap via condensation. Because the sample is not diluted prior to counting, very low concentrations can be measured. High sensitivity HTO-in-air measurements may also be made by cooling the air in a cold finger and collecting the condensate. However, these high sensitivities are not generally required for radiological protection.

Another technique for quickly monitoring for HTO in air is to employ a soft plastic bottle which is squeezed several times in the area where the measurement is to be made. Once the HTO concentration in the bottle is approximately representative of the room concentration, a measured amount of clean water is quickly introduced into the bottle, the bottle shaken and the water later counted for HTO. From the concentration of HTO in the water, the concentration of HTO in room air can be calculated. A method for calculation is given in Ref. [41].

### 3.3.2. Ionization chamber methods

The most common real time method of measuring tritium in air is with an ionization chamber (Fig. 8). Because of its low  $\beta$  energy, tritium has to be introduced directly into the ionization chamber. Ion pairs produced in the measuring chamber from the tritium  $\beta$  particles are collected by an electric field applied across the chamber. Ion pairs produced outside the measuring chamber must be removed before they can enter the chamber by a particulate filter and an ion precipitator. It is important that the filter material not significantly trap HTO. Charcoal, for example, should not be used.

Because of the relatively low number of ion pairs produced per tritium  $\beta$  particle, individual pulses are not measured. Instead, the chamber is operated in the current mode where the current is proportional to the tritium activity. As an approximate rule of thumb, the ionization current from tritium is  $1 \mu\text{A}$  per  $40 \text{ GBq}$  ( $1 \mu\text{A}/\text{Ci}$ ). Thus, for a  $1 \text{ L}$  ionization chamber and a tritium concentration of  $40 \text{ kBq}/\text{m}^3$  ( $1 \mu\text{Ci}/\text{m}^3$ ), the ionization current is about  $1 \text{ fA}$ . A very sensitive and stable electrometer with associated electronics is required to achieve this sensitivity. It is also important that insulating surfaces be kept scrupulously clean. A tritium-in-air concentration of  $40 \text{ kBq}/\text{m}^3$  ( $1 \mu\text{Ci}/\text{m}^3$ ) is about the practical lower limit of tritium concentration measurable by a small ionization chamber in the field.

A major difficulty with ionization chambers is that they are not specific to tritium. They respond to other airborne radioactive substances as well as to ambient  $\gamma$  radiation. For example, background radon may interfere with attempts to measure tritium concentrations of a few tens of  $\text{kBq}/\text{m}^3$  in air. The ionization current from a  $\gamma$  exposure rate of  $1 \text{ mR}/\text{h}$  ( $0.258 \mu\text{C}\cdot\text{kg}^{-1}\cdot\text{h}^{-1}$ ) at sea level is the same as that from a tritium concentration of  $5 \text{ MBq}/\text{m}^3$ .

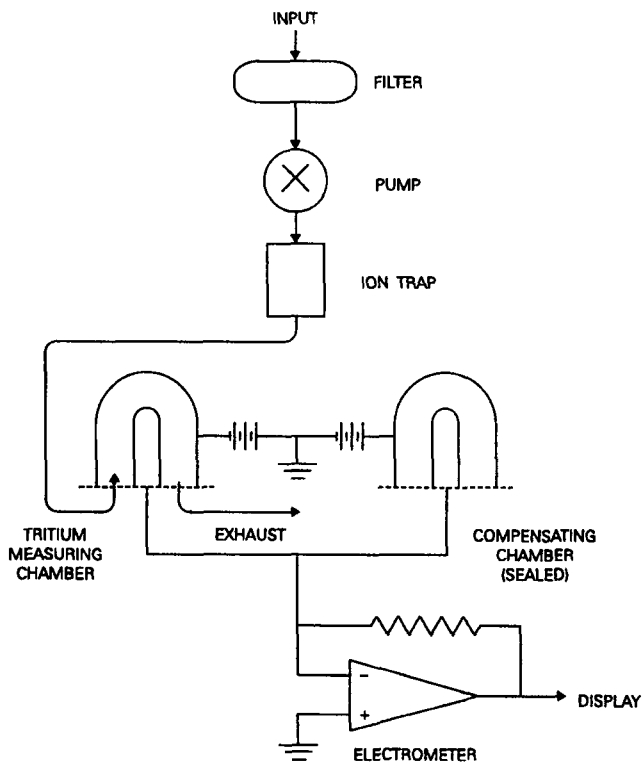


FIG. 8. Ionization chamber monitor.

To reduce the response to  $\gamma$  radiation, compensating chambers are commonly used. The tritium sample flows through only one chamber, while the  $\gamma$  radiation affects both chambers. The signal due solely to the tritium is the difference in ionization current between the two chambers.

In practice, the best that can be achieved with simple compensation schemes is about 97% suppression of the  $\gamma$  signal, owing to inherent geometry effects and to changes in the response of the tritium measuring chamber to changing atmospheric conditions if the compensating chamber is sealed and cannot respond to these changes.

Many different physical arrangements of the two chambers have been tried. Ideally the compensating chamber is identical to the tritium measuring chamber. The fundamental difficulty is that for perfect cancellation, the two chambers must occupy the same position in space at the same time — a physical impossibility. The most common arrangement is for the two chambers to be mounted side by side or base to base. This arrangement works quite well in homogeneous  $\gamma$  fields, but the

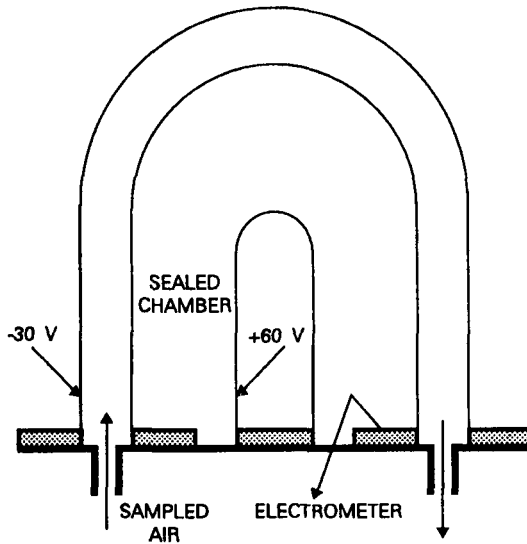


FIG. 9. Ionization chamber tritium monitor with  $\gamma$  compensating chamber.

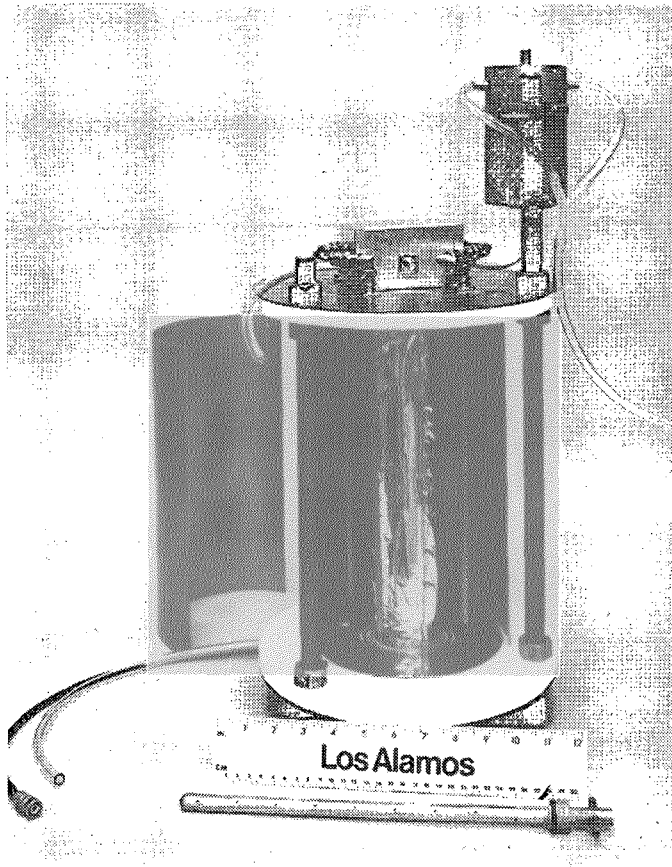
presence of point sources close by can give rise to inaccuracies. The resultant error is not always conservative.

A concentric arrangement [42] of the ionization chambers provides better compensation in an inhomogeneous  $\gamma$  field but at the price of added complexity and cost (Fig. 9).

A more complicated method is to mount the two chambers on a rotating turntable [43]. Thus each chamber occupies the same position in space, but not at exactly the same time. This monitor is capable of detecting about  $500 \text{ kBq/m}^3$  ( $10 \text{ } \mu\text{Ci/m}^3$ ) in a steady or slowly changing  $\gamma$  field of  $500 \text{ } \mu\text{Gy/h}$  ( $50 \text{ mR/h}$ ) after several minutes of observation.

Another scheme that has been reported switches a clean air flow and the sample flow back and forth between two identical chambers [44]. This results in excellent  $\gamma$  suppression but at the cost of a long time response (several minutes).

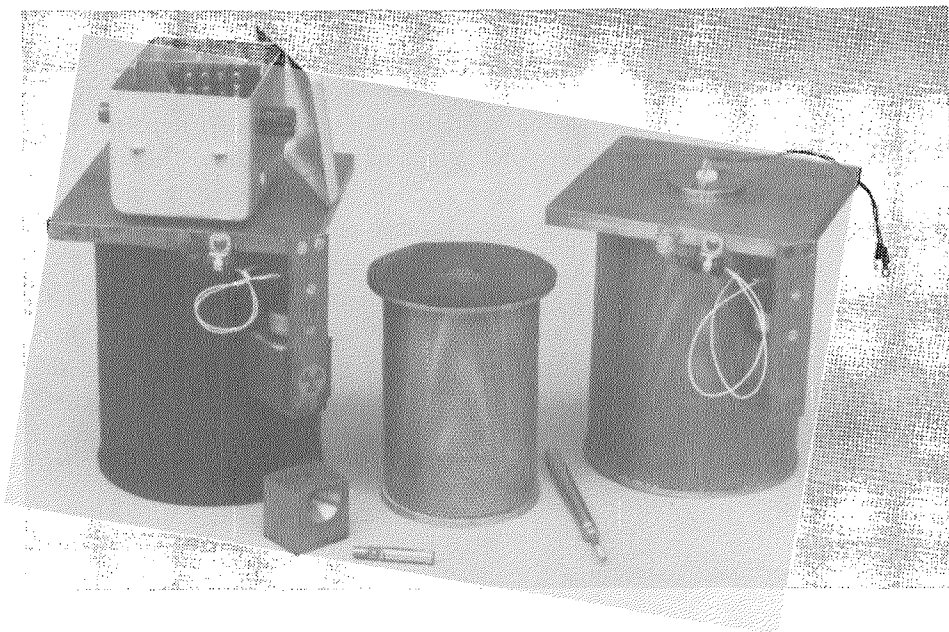
The ionization current from a given concentration of most common  $\beta$  emitters is greater than that from the same concentration of tritium. However, these ionization currents are usually less than one would predict from a simple scaling of the  $\beta$  energy. This is because, while a particular ionization chamber may be large compared with the range of a tritium  $\beta$ , it usually is not large compared with the ranges of other  $\beta$  emitters. Not all of the  $\beta$  energy is lost before the  $\beta$ 's strike the wall. As a result, the counting efficiency for a particular radionuclide is geometry dependent and varies from chamber to chamber.



*FIG. 10. Concentric double ionization chamber for discriminating between tritium and other gaseous  $\beta$  and  $\gamma$  emitters.*

The presence of other radioactive gases may mask the presence of a much larger hazard due to HTO. For measuring tritium in the presence of other  $\beta$  emitting radioactive gases, Jalbert [45] has developed an effective variation of the compensating chamber scheme. Concentric cylindrical ionization chambers share a common wall of a screen material covered by aluminized Mylar or thin aluminium foil opaque to tritium  $\beta$ 's. The sample gas enters the inner measuring chamber through a hollow central electrode (to minimize transient effects). The tritium is measured in the inner chamber, whereas the high energy  $\beta$ 's from the contaminant gas produce current in the inner chamber and penetrate the common wall to the outer chamber where electrodes collect the compensating current (Fig. 10).





*FIG. 11. Ionization chamber with perforated walls.*

For measuring HTO in the presence of other radioactive gases, a more common scheme is to split the sample flow in two, with one branch going directly to a measuring chamber while the second stream passes through a desiccant before entering a second chamber. The tritium concentration is then obtained by current subtraction. The flow switching monitor previously mentioned in the context of  $\gamma$  discrimination is a variation of this technique [44].

The more specialized problem of measuring HTO in the presence of HT is discussed in more detail in Section 3.3.4. The techniques mentioned there can also be adapted for suppressing the response to other radioactive gases.

For measuring very high ionization currents, care must be taken in the electrode design. If the collection volume has low electric field regions, there may be significant non-linearity due to recombination effects [46]. While a flat parallel plate geometry produces a very efficient collection owing to the uniform electric fields, cylindrical geometries are much more common. Ideally, the central collecting electrode of such a chamber should be of large diameter to counteract the geometry problem.

Other difficulties associated with ionization chambers are that they are subject to contamination problems (memory effects), are sensitive to vibration and can often be put completely out of action by a prolonged period of high humidity. Condensation on the insulating surfaces can destroy the isolation resistance of the collection

electrode. The use of Teflon or Kel-F insulators will generally minimize some of these problems.

Contamination of ionization chamber surfaces by HTO can be reduced by the use of highly polished surfaces. The surface finish appears to be more important than the particular metal used. It is also helpful to minimize the wall area. Thus the measuring chamber walls may be made of screen, grid or perforated sheet material. This has the added benefit of reducing interactions with  $\gamma$  and X ray fields and hence the background.

Ionization chambers usually require a pump to move the air sample through the measuring volume. For glove boxes and room exhaust ducts, an open mesh ionization chamber can be used to provide a measure of the tritium activity in the air. Ionization chambers have also been used for simple room monitoring. An example is shown in Fig. 11. This design eliminates the need for mechanical pumps and reduces ionization chamber contamination.

### 3.3.3. Proportional counters

In proportional counters the tritium in the air is mixed with a counting gas such as argon-methane and is introduced into the counter. The number of ion pairs produced is increased by gas amplification in the strong electric fields surrounding the anode. The amplification is sufficient to produce a detectable charge pulse. The number of pulses (of appropriate energy) is proportional to the tritium concentration. By using multiwire chambers and pulse shape or pulse height discrimination, it is possible to discriminate between tritium  $\beta$  particles and  $\alpha$  particles or electrons produced by  $\gamma$  radiation.

In principle proportional counters can offer much higher sensitivity to tritium than ionization chambers. However, in practice this higher sensitivity is often hard to realize. For small samples of high specific activity HTO or for some other tritiated species such as HT or tritiated methane the higher sensitivities can be achieved. However, atmospheric air is deleterious to most counting gases. This limits the amount of sample that can be added. The review of tritium measuring techniques by the National Council on Radiation Protection and Measurements (NCRP, USA) [32] lists the sensitivities that can be achieved by the various measurement techniques.

Proportional counters are used in countries where regulations require the monitoring of very low concentrations below the range of ionization chamber instruments. Measurement of tritium concentrations of  $500 \text{ Bq/m}^3$  ( $0.01 \mu\text{Ci/m}^3$ ) is achievable with a counting time of about one minute.

Proportional counters are of higher cost, are more complex and require a counting gas (usually methane or filtered natural gas). The upper range of such instruments is limited (typically  $50 \text{ MBq/m}^3$ ). However, such instruments have an inherent ability to discriminate against external penetrating radiations or radiation

from other radioactive gases accompanying the tritium. They are particularly suited to stack monitoring where the tritium concentrations are generally expected to be low.

#### 3.3.4. HT-HTO discrimination

At the same concentration in air, HTO is about four orders of magnitude more hazardous than HT. Tritium monitors with discrimination ratios of at least this magnitude are necessary if the hazard is not to be overestimated when the ratio of HT to HTO is large.

It is unlikely, however, that large HT:HTO ratios will be found in non-accident situations. Considerations other than the direct radiological hazard to workers will likely limit the chronic HT-in-air concentrations. For example, derived release limits are often not species specific [47] and are generally based on total tritium. To meet these release limits it is necessary to control the HT concentrations more strictly than would be required if only the immediate radiological hazard to the workers were being considered. While differentiating between HT and HTO is desirable in principle, it may not be necessary in the normal day to day operation of the facility.

Ontario Hydro has decided that no discrimination is required for its Darlington Tritium Removal Facility [48]. The rationale for this decision is as follows: (1) all tritium is assumed to be HTO, which is the conservative assumption; (2) the chronic HT levels are expected to be low; (3) HT is eventually oxidized to the more hazardous HTO in any case; and (4) there is a physical separation between those areas where HT predominates and those areas where HTO is the more prevalent. In general, facilities that handle large but well contained quantities of HT have adopted this approach for similar reasons.

On the other hand, the Chalk River Nuclear Laboratories have chosen to provide a species specific tritium monitor for their tritium extraction plant so that a given hazard can be quickly and accurately assessed. This monitor will also provide useful data on the actual HT:HTO ratios encountered in an operating tritium facility. The monitor is based on the use of a permeable membrane to separate the HTO from the sample stream [49].

For maintenance activities a discriminating monitor may be useful. Timely determination of the actual concentrations of HT and HTO present can facilitate efficient and safe disassembly of tritium process and containment systems.

The effective dose equivalent resulting from inhaled HT is due mainly to lung irradiation and can best be estimated by air monitoring. In the absence of an air monitoring programme a conservative course would be to multiply the doses estimated from tritium in urine by two (see Section 2.3). However, it is only when the ratio of HTO to HT is very low that the dose to the lungs will be significant compared with the whole body dose due to HTO (regardless of origin). For example, when the concentration of HTO in air is 1/1000 of the HT concentration, the lung dose from

HT will contribute 10% to the effective dose equivalent [16]. Such low HTO:HT ratios are unlikely to be found in non-accident situations.

For accident situations, a judgement is necessary as to whether or not an explicit determination of the actual — as opposed to the potential — concentrations of HT and HTO is required, and of how much time can be allowed for this determination. For accident situations a gross overestimation of the hazard is not necessarily prudent. Overestimating hazards can give rise to inappropriate responses (such as evacuation) that may actually increase the risk. Bubbler (or passive sampler) measurements combined with measurements of total tritium in air can accurately determine the actual concentrations of HT and HTO, but the turn-round time for the measurements is unlikely to be less than an hour. If a rapid response is necessary, a real time monitor which is calibrated for specific compounds of tritium will have to be provided. Various methods can be considered.

The most straightforward method of measuring HTO in the presence of HT is to measure the total concentration followed by a second measurement of the air after passing it through a bubbler or desiccant. The HT concentration is determined from the second measurement while the HTO concentration is determined from the difference between the two measurements. Such methods have previously been used to measure the concentration of HTO in the presence of other radioactive substances such as radioactive noble gases. This method is appropriate for most practical situations where the HT:HTO ratios are not too high, but it is not appropriate for HT:HTO ratios of the order of 10 000. There is no practical method to determine the difference between two tritium concentrations which differ by only 1 part in 10 000.

Bubbling a gas sample flow through water and then purging with clean air is an effective way to separate HTO from other radioactive gases. The HTO remains in the water. Osborne [50] developed a real time HTO-in-air monitor based on this method. This monitor provides good sensitivity to HTO, good immunity to the presence of other radioactive gases and reasonable immunity to  $\gamma$  radiation. Its response to HT is expected to be of the order of  $10^{-3}$ – $10^{-4}$  of the response to HTO.

Some materials are much more permeable to water vapour than to other gases. Howell et al. [51] and Beach and Hoots [52] developed monitors based on the diffusion of HTO through dimethyl silicone. These monitors used a proportional counter as the detector and gave an output signal proportional to the combined hazard of the HT and HTO present (according to the previous ICRP recommendation of a relative hazard of 400 to 1 between HTO and HT). To obtain a discrimination ratio of 400 (i.e. to be 400 times as sensitive for HTO as for HT), two permeation stages were necessary. In principle, discrimination ratios of the order of 10 000 could be achieved by using more permeation stages, but at the cost of reduced sensitivity and increased response times.

Another material with a good permeability to water vapour is Nafion, a material developed by Du Pont. The permeation rate of HTO is more than  $10^4$

times greater than that of HT [53]. This material can be used to construct tritium monitors with discrimination ratios closely approaching those required. The major problem with this method is the response time, an inherent characteristic of the diffusion process. Through careful design some improvement is possible. Nafion based monitors have been reported in Refs [49, 54–56] (Fig. 12).

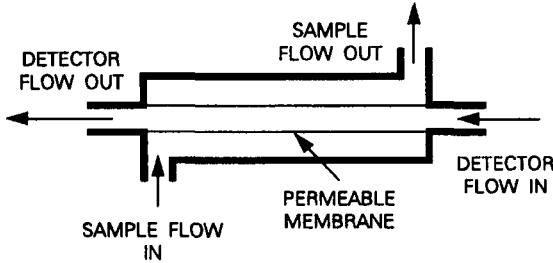


FIG. 12. Nafion based tritium monitor.

### 3.3.5. Calibration

Ideally a tritium monitor should be calibrated with the same compound that it is designed to measure. For example, HTO-in-air monitors should be calibrated using HTO. This will ensure that surface effects are properly taken into account. The retention of HTO on ionization chamber surfaces can, in some circumstances, affect the calibration by up to 10%. This implies that HTO calibrations are humidity dependent, and that indeed is the case.

A common procedure for calibrating an HTO monitor is to produce a stream of tritiated air by passing air through a temperature controlled bubbler containing tritiated water. The temperature of the bubbler and the specific activity of the water in the bubbler can be adjusted to give a wide range of HTO-in-air concentrations and relative humidities. The tritiated air stream is passed through the monitor and subsequently collected in two bubblers in series. The tritium collected in the bubblers is measured and used to determine the exact concentration that passed through the monitors. A thorough discussion of this type of calibration procedure is given by Banville [57].

A more common calibration method for ionization chamber monitors is based on the use of known concentrations of HT or  $\text{CH}_3\text{T}$ . Known aliquots of the gas are injected into the monitor's chamber arranged in closed loop fashion. Once a stable, reliable monitor is calibrated, it can be used to calibrate other monitors by connecting them in a closed loop and injecting less expensive tritium gas whose precise concentration need not be known. Because HT will be adsorbed on the walls of the calibration loop, the loop should be flushed with HT several times before calibration. A continuous stream arrangement is preferable.

Simple operational checks are most easily performed using solid  $\gamma$  sources. This is not a substitute for a true calibration, however.

### **3.3.6. Tritiated dust**

The machining or abrading of tritium contaminated materials may give rise to tritiated aerosols. The handling of powders containing tritiated materials (tritiated pharmaceuticals, tritiated thymidine, etc.) can also present a hazard to workers. In all these cases air sampling can be carried out with a filter paper sampler and pump, using either an installed area sampler or preferably, where the risk is significant, personal air samplers. The filter paper samples can be counted in a liquid scintillation counter, with a solid scintillator or in a  $2\pi$  proportional flow counter.

## **3.4. TRITIUM-IN-LIQUID MONITORING**

### **3.4.1. General discussion**

Tritium-in-water monitoring is often necessary for the control of tritium releases to the environment and as an early indication of process system leaks. It is sometimes desirable that these measurements be performed continuously with an on-line monitor. The required sensitivity and time response will depend upon the purpose of the monitoring and the particular situation. However, it is often desired to measure down to several tens of kilobecquerels per litre.

Currently, most tritium-in-water monitoring is by grab sampling or, rarely, by some type of on-line scintillation counter. Neither of these methods is entirely satisfactory. The problems of on-line tritium-in-water monitoring have been discussed in Ref. [58].

### **3.4.2. Grab samples**

The most common, direct and straightforward method of measuring the tritium concentration in water and other liquids is to take a grab sample of the liquid and measure its tritium concentration using liquid scintillation counting techniques. The method is very sensitive but does not give results in real time. In spite of this disadvantage, this method is the most reliable method of measuring tritium in water. It is widely used and is adequate where the required sampling rate is not high.

### **3.4.3. Liquid scintillation counting**

Liquid scintillation counting is a basic technique that is used in many different aspects of tritium work. The field is quite specialized, with a large literature of its

own. It is not appropriate to fully cover that work here. A detailed discussion of liquid scintillation techniques is given in Refs [59–61] and more recent developments are reported in Refs [62, 63].

In liquid scintillation counting, a scintillation solute is dissolved in a suitable solvent to produce a liquid scintillation cocktail. The solvent should be miscible with the liquids to be monitored for tritium. An aliquot of the liquid to be measured is mixed with the liquid scintillation cocktail. The scintillations produced by the energy deposition from the tritium  $\beta$  particles are detected by two photomultiplier tubes, operated in coincidence, to reduce noise at the high sensitivities required. Tritium levels down to 400 Bq/L can be measured in most liquids.

Several automated liquid scintillation batch sampling systems have been described [64–66]. These systems usually consist of a system that mixes liquid scintillator and a sample of the water to be measured and then counts the mixture in a standard liquid scintillation counter. It is necessary to ensure that the water and scintillation cocktail are properly mixed. A major problem with these systems is to provide a clean sample. Samples often contain materials that can lead to ‘quenching’ and sometimes chemiluminescence.

#### **3.4.4. Scintillation flow cells**

Several continuous tritium-in-water monitors have been developed using flow cells containing solid scintillators such as anthracene coated Plexiglas [67], scintillator plate [68], thin scintillator sheets [69], scintillator sponge [70], scintillator film [71], doped quartz balls [72] and plastic scintillator beads [73]. Plastic scintillator capillary tubing and glass tubing filled with anthracene crystals embedded in a clear plastic flow cell are commercially available. The latter is the more sensitive and the crystals may be replaced if contaminated. Plastic scintillators exposed to tritium can build up a background with time that is difficult to eliminate.

Purging the water sample by bubbling a clean, non-radioactive gas through it is an effective method of reducing the concentration of radioactive gases dissolved in the sample. Appropriate filtration can reduce the effects of radioactive particles. Lead shielding can reduce the effect of external  $\gamma$  radiation.

Most of the monitors mentioned above are sufficiently sensitive for measuring HTO concentrations of a few tens of kilobecquerels per litre in laboratory conditions. However, these monitors often fail in the field. Effluents requiring monitoring are often dirty, and filtration down to at least 5–10  $\mu\text{m}$  pore size is necessary [74]. Even at this size, some dirt (silt, charcoal particles, slime and algae) in the sample water can pass through the filters and foul the surface of the scintillator. In addition to dirt and other radioactive materials, luminescent material and chemiluminescent material (chlorine, for example) are often present.

### 3.5. SURFACE CONTAMINATION MONITORING

#### 3.5.1. General discussion

In actual situations the contamination will not only be 'on' the surface but will also penetrate 'into' the surface. For most contaminants on most surfaces the penetration will be small, but for tritium it will almost inevitably be large compared with the range of the tritium  $\beta$  particle. Thus tritium atoms that are associated with the surface may not contribute to any 'surface activity'.

There are three different types of tritium surface contamination. 'Total tritium surface contamination' is the total amount of tritium adsorbed on and absorbed into the surface. 'Directly measurable tritium surface contamination' is the fraction of the contamination that can be measured directly by a radiation detection instrument held close to the surface. 'Removable tritium surface contamination' is the fraction of contamination that can be removed by wiping or washing.

Total tritium surface contamination is not readily measurable without destroying the surface. Leaching can be effective, however, for removing tritium from metal surfaces for subsequent measurement, especially if the surfaces have not been exposed to tritium for too long. Directly measurable tritium surface contamination is measurable, in principle, by real time instruments such as open window proportional counters. A fraction (often taken to be 10%) of the removable surface contamination is measurable by smears.

The immediate risk to workers from tritium surface contamination is believed to correlate with the removable fraction of the contamination. Thus for radiological protection purposes, measurement of the removable surface contamination is appropriate. It must also be remembered, however, that the removable contamination can be replenished from the tritium absorbed beneath the surface. For certain purposes, such as the disposal of wastes, measurement of the total tritium contamination would be more appropriate if it were possible.

There have been various studies of methods to assess tritium surface contamination and the resultant radiological hazard [75-78]. The most important of these is the work that was done at the Atomic Energy Research Establishment, Harwell, United Kingdom, in the late 1960s and early 1970s. This work included studies of the sorption of tritium on brass [79], the estimation of tritium contamination by smearing [80] and the radiological hazard [18]. These data were used by Johnson and Dunford [20] to evaluate various dosimetric models pertaining to percutaneous absorption of tritium from surfaces. This analysis showed that significant skin doses can arise from contact with metal and other surfaces that have been exposed to high concentrations of  $T_2$ .



### 3.5.2. Smear techniques

While there has been a continuing interest in the development of real time instruments for the measurement of surface contamination [75, 81–85], the smear technique seems to be accepted as the implicit standard method for evaluating the hazards from tritium surface contamination.

Real time instruments are subject to contamination problems, especially if the surface is highly contaminated or outgassing rapidly. Also, many probes are designed only for smooth, flat surfaces and may require a counting gas (proportional or Geiger–Müller probes).

A suitable smearing (swipe) procedure is as follows. The surface to be measured is uniformly wiped with a dry or moist piece of swipe material (filter paper or blotting paper, for example). The total area swiped must be known or estimated. A few tens of square centimetres is appropriate but 100 cm<sup>2</sup> is typical. The swipe is then introduced into a standard liquid scintillation vial that has been previously filled with liquid scintillation cocktail. The vial is allowed to stand 10–20 min before counting in a liquid scintillation counter. A typical removal efficiency is 10% but large variations are possible. The counting efficiency will have to be determined for the particular combination of liquid scintillation cocktail and swipe material used. The effect on counting efficiency can be as much as a factor of 2.

The choice of scintillation cocktail does not appear to be particularly critical. Wet swipes (wetted with water or glycerol) are sometimes used for higher sensitivity and reproducibility. However, adequate sensitivity is available with dry swipes, but significant losses may occur if the paper is not quickly inserted into the liquid in the counting vial. At Ontario Hydro a ‘Poly foam’ smear that is soluble in the scintillation cocktail has been found to be convenient and satisfactory.

As an alternative to liquid scintillation counting, swipes are sometimes measured in proportional flow counters or plastic scintillation counters. This has the advantage that the swipe may be measured in the field but losses of tritium and contamination of the counters can occur.

Uncertainty in estimates of the total tritium surface contamination based on swipes can be very large. In spite of this, swipe measurements are likely to be reasonable predictors of the radiological hazard, assuming that the hazard correlates with the removable tritium surface contamination.

## 3.6. TRITIUM-IN-SOLIDS MONITORING

The direct determination of the total tritium activity in solid wastes is almost impossible. The removable surface contamination and the emanation rate can be

measured. The latter may be influenced by temperature, humidity, surface conditions and other factors. Although the safety of handling the material can be determined, it is not feasible to determine the total tritium activity in solids except by complete destruction or dissolution of the material.

## **4. PERSONAL PROTECTIVE CLOTHING**

### **4.1. INTRODUCTION**

This section provides guidance on the selection and use of personal protective clothing for work in tritium contaminated atmospheres and for handling tritium containing or tritium contaminated equipment and materials. Personal protective clothing refers to clothing and equipment worn directly by the worker. Protective equipment and techniques such as glove boxes, fume hoods and ventilation control are discussed in Section 7.

The objectives of personal protective clothing are:

- (a) To reduce the uptake of tritium by inhalation, skin absorption or ingestion;
- (b) To prevent the occurrence of significant levels of contamination on personal clothing or skin;
- (c) To prevent the spread of contamination from contaminated to clean areas.

Protective clothing that achieves these objectives includes smocks or coveralls, gloves, booties, respirators and full body plastic suits.

### **4.2. SMOCKS AND COVERALLS**

In a room where contamination levels are low or are generally confined to equipment in fume hoods and glove boxes, a minimum of protective clothing may be all that is required. Laboratory coats or smocks are quite adequate while working in such a room but not actually handling contaminated equipment. In lieu of smocks, coveralls may be necessary if the former do not offer adequate protection for the work to be done. Such clothing is generally made of cotton and is reusable after washing. Disposable smocks or laboratory coats are also commercially available. Generally they are made of paper but also may be constructed of laminated plastic materials for greater protection against HTO. However, the latter materials may be uncomfortable in warm working environments.

### 4.3. GLOVES

When handling material or equipment that is contaminated or potentially contaminated, workers should wear gloves to minimize the uptake of tritium through the skin.

Gloves should offer sufficient protection against tritium permeation while providing reasonable comfort. They should also allow for reasonable dexterity and be acceptably long-wearing and resistant to chemicals. They are generally inexpensive and are usually discarded after each use in order to reduce the spread of contamination. Such gloves are generally made of latex or polyvinyl chloride (PVC). Polyvinyl chloride is usually preferred because of its lower permeability to HTO and HT and its resistance to chemical attack. Disposable latex gloves have limited shelf life at many facilities owing to deterioration caused by ozone. In addition, many of the disposable latex gloves may be plagued with quality control problems due to pinholes that result from the manufacturing process.

If rough work is to be performed, additional gloves or heavier gloves should be worn. A good solution may be to wear a heavy glove on the inside with a less expensive glove on the outside. The disposable glove may be discarded after the job and the more expensive glove used again if it is not damaged or contaminated. Minimum thicknesses recommended depend on the contamination levels and the time between glove changes. Generally gloves of 0.5 mm thickness are quite adequate for handling surfaces with removable contamination up to 5 GBq/m<sup>2</sup> (1 mCi/100 cm<sup>2</sup>) or more if the gloves are changed frequently (every ten minutes or so).

Laboratory rules and common sense dictate that workers remove contaminated gloves before they touch any uncontaminated surface. The technique involves pulling down the wrist of one glove about two thirds of the way with the other, gloved, hand, and removing the second glove completely with the still covered fingers of the first hand. Both gloves should be discarded into the radioactive waste container.

Most disposable gloves are short, wrist length gloves. Elbow length or even longer gloves should be used when handling highly contaminated equipment that may come into contact with the arms, and when using the gloves of boxes with high tritium concentrations (> 5 GBq/m<sup>3</sup>). Two or three sets of gloves may be worn when working with high levels of tritium contamination; for example, the inner glove may be cotton (for comfort), the second, polyethylene (for impermeability) and the outer surgeon's latex gloves (to protect the upper arm).

Experience and specific working conditions will dictate the rules or requirements for wearing gloves in any particular situation. The chemical compatibility of glove materials and the physical properties of commercially available gloves are given in Tables III and IV.

TABLE III. CHEMICAL COMPATIBILITY OF AVAILABLE LIQUID-PROOF GLOVE MATERIALS

Material	Recommended for:	Not recommended for:
Butyl	Dilute acids and alkalis, ketonic solvents, gas and vapour permeation protection	Petroleum oils, distillates, solvents
Natural rubber	Ketonic solvents, alcohols, photographic solutions	Petroleum oils, distillates, solvents
Neoprene	Concentrated non-oxidizing acids, concentrated alkalis	Halogenated and ketonic solvents
Neoprene-natural rubber blends	Dilute acids and alkalis, detergents, photographic solutions	Halogenated and ketonic solvents
Nitrile	Petroleum based solvents, distillates, oils	Halogenated and ketonic solvents
PVC	General purpose, low risk hand protection	Halogenated and ketonic solvents
PVA	Halogenated solvents, paint shop applications	Water and water based solutions
Viton	Halogenated solvents, concentrated oxidizing acids	Aldehydes, ketonic solvents

#### 4.4. BOOTIES OR SHOE COVERS

If the floor of the tritium handling room is actually or potentially contaminated, booties may be required. Very often, in laboratories where tritium is confined to glove boxes and hoods, contamination on floors, walls and other surfaces is virtually eliminated and booties generally are not required. In some rooms and laboratories, however, booties may be required. A good surface contamination monitoring programme is essential to any decision making regarding the type of protective clothing that would be appropriate.

TABLE IV. COMPARISON OF COMMERCIALY AVAILABLE GLOVES

Material	Thickness (cm)	Shelf life	Relative toughness	HTO permeation characteristics
PVC	0.013	Fair	Fair	Poor
PVC	0.025	Good	Good	Fair
PVC	0.05	Excellent	Excellent	Good
Neoprene-natural rubber blend	0.05	Good	Good	Good
Neoprene	0.038	Excellent	Good	Good
Neoprene	0.056	Excellent	Good	Good
Natural rubber	0.038	Poor	Fair	Good
Nitrile	0.038	Excellent	Excellent	Good
Nitrile	0.056	Excellent	Excellent	Good
Butyl	0.03	Excellent	Poor	Excellent
PVA <sup>a</sup>	0.056	Good	Excellent	Poor
Viton	0.03	Excellent	Excellent	Excellent

<sup>a</sup> The thickness of PVA gloves fabricated from a coated, flock lined fabric can vary by as much as 20%. Because the PVA coating is water soluble, the properties of PVA gloves can be expected to vary depending on their long term exposure to moisture.

#### 4.5. AIR SUPPLIED BREATHING APPARATUS

Many circumstances arise where the principal hazard is the possibility of a sudden release of airborne HT directly into the breathing area of the workers. The amount of tritium involved may not be great, or the probability of a release, often determined by experience, may be small. Complete skin protection may not be necessary, since high air concentrations would be short lived and, if not, the workers would have adequate time to retreat from the area. In such cases, although the use of smocks and gloves alone may be insufficient, full body plastic suits might not be appropriate.

Under such conditions air supplied breathing apparatus in addition to smocks (or coats) and gloves would be appropriate. Such apparatus consists of a partial or (usually) full face mask supplied with air (demand or continuous) from a building

air supply or from an air tank carried on the back (self-contained breathing apparatus (SCBA)). The advantage of such equipment over the more complete protection afforded by full plastic suits is that workers can more readily put it on and can perform their tasks more comfortably, thereby lessening the possibility of mistakes that can result in an accidental release. SCBAs are available from several suppliers. A report describing several of the types used in the United States of America and in Europe has been published [86]. It was found that the pressure demand type of respirator offers a greater degree of respiratory protection ( $10^3$ – $10^4$ ) than does the demand type ( $10$ – $10^3$ ). This is not unexpected, since the demand type receives clean air only when the wearer inhales and produces a negative pressure. Tritium leaking into the respirator around the seal has a significant probability of reaching the breathing zone during inhalation. In the case of the pressure demand SCBA, a positive pressure is maintained in the face-piece at all times, thus helping to prevent any tritium inleakage into the face-piece. In the USA, SCBAs are typically designed to meet the requirement for a 30 min wearing time under average conditions.

For protection from exposure to HT the protection factors mentioned above are applicable since permeation of HT through the skin is very slow and the lung is considered to be the organ at risk. In the case of exposure to HTO, however, the HTO readily diffuses through human skin, and so respirators offer only partial protection. For a person breathing at a rate of approximately 20 L/min (light work) the intake of HTO into the body is roughly divided between the lungs (67%) and the skin (33%). For long exposures the best respirators can then offer a total protection factor of only 3. For very short exposures, however, as in the case where the SCBA is worn for emergency protection, the total protection factor may approach that of the respirator because superficial skin contamination may be quickly washed off and ordinary clothing provides at least some delay to the movement of tritium toward the skin.

In cases where the airborne tritium hazard is low, in areas where there is no air supply or where the worker has to be mobile and cannot be attached to the air supply header for a long period, a self-contained full or partial respirator may be used to good advantage instead of an air supplied respirator. The respirator may be equipped with a cartridge filled with desiccant which removes the HTO. However, the inspired air could be too dry and create discomfort. One type developed by Ontario Hydro for use at its CANDU stations [27] consists of a canister type air purifier filled with exfoliated vermiculite which is wetted with water before use. HTO is removed by isotopic exchange. Tritium breakthrough after one hour of use is about 5%.

#### 4.6. AIR SUPPLIED SUITS

HTO vapour diffuses through the skin, resulting in an internal dose that is comparable to that from inhalation when a person is exposed to airborne HTO. Therefore

the use of ventilated plastic suits completely enclosing the body is necessary for effective protection in high concentrations of HTO in air. Sometimes air supplied suits may be used to advantage in the handling of large, highly contaminated equipment, to guard against tritium uptake through skin contact. In general, however, it is preferable to protect personnel by enclosing the equipment rather than requiring the wearing of cumbersome suits.

HT in the gaseous state is not absorbed by the skin to a significant extent and so in principle only protection against inhalation is necessary. In the work environment, HT is converted to HTO to some extent. If the airborne tritium concentration is high and the relative concentrations of HTO and HT are not known, it would be prudent to assume all tritium to be HTO and to use plastic suits for protection. If the relative concentrations of HTO and HT are known, for example by the use of HTO/HT discriminating monitors, it would be possible to decide whether simple respiratory protection would suffice.

In general plastic suits hinder the wearer, so that a given task will take longer to perform, typically 10 to 50% longer. If the work is done in high external  $\gamma$  fields, this extra working time could result in additional external dose that exceeds the savings in tritium dose. In such situations a judgement will have to be made as to whether to rely on respiratory protection alone, or even to have no protection.

Air supplied suits must provide adequate protection against tritium intakes by both inhalation and skin absorption. The suits should be properly designed and well fabricated, using materials that are resistant to tritium permeation. The air distribution system provides fresh breathing quality air for inhalation and body cooling, as well as a positive pressure against the ingress of contaminated air. Protection factors (the ratio of tritium uptake without suit protection to uptake with suit protection) in excess of 1000 can be achieved when strict usage procedures are followed. Such protection factors have been measured under simulated work conditions in the laboratory [27]. In practice the protection factor is lower and depends on suit design as well as usage procedures.

Air supplied suits come typically in one, two or three pieces, consisting of a jacket, pants and possibly a detachable hood. Typically the jacket has hard cuffs on the ends of the sleeves to which gloves can be attached. The booties are normally welded to the pants, making the pants and booties one piece. Additional booties may be worn if the floor is highly contaminated or if the built-in booties are not designed to be worn without additional booties. Breathing air is supplied generally to the hood and flows out of the suit through the bottom of the jacket, or through exhalation ports provided at strategic locations to assure a flow of uncontaminated air throughout the suit. Ventilating and cooling air may be distributed to the arms and legs of the suit through an air distribution harness which is worn under the suit.

In the USA several government contractors and laboratories have developed their own air supplied plastic suits. Many of these were developed for handling other hazardous materials and later some were adapted for tritium work. Most are of the

two piece design and constructed of PVC 6–12  $\mu\text{m}$  thick. Exceptions to this are the two suits recently developed by the Savannah River Plant. One uses 8  $\mu\text{m}$  laminated Tyvek (spin bonded polyolefin), PVC and saran/chlorinated polyethylene, and the other is constructed of 7  $\mu\text{m}$  polyurethane film. The saran provides an additional barrier to penetration, especially by HTO, and the polyurethane provides additional resistance to tearing and punctures.

In Canada most air supplied suits in use are those developed by Ontario Hydro for use at its CANDU stations. Currently a two piece suit is used which incorporates a one piece reusable body section with arms, legs and attached booties, plus a separate disposable hood. The body section has air supply channels and a communication interface built in as an integral part of the suit. The suit is designed for user comfort and for quick, unaided dressing and undressing, which are important requirements at CANDU stations. The suit material for the body section is made of PVC. A dry-cleanable version of the suit is made of a neoprene-on-nylon material. The disposable hood is made from Number 1422 Tyvek laminated with 0.044 mm thick polyethylene.

Suit materials must have low tritium permeability, particularly for HTO, and must be sufficiently tear and abrasion resistant to stand up to field use. They should be non-flammable, non-allergenic, inert to common chemicals (especially oils and grease), relatively easy to fabricate and comfortable to wear.

The air flow required for each suit depends on the suit design and the operational requirements of its intended use. Suits used in the USA typically have design flow rates of 0.2–0.6  $\text{m}^3/\text{min}$  at a recommended air supply header pressure of about 500 kPa gauge. In Canada a flow rate of 0.8  $\text{m}^3/\text{min}$  at a supply pressure of 515 kPa [27] is normal. The higher flow rate is considered necessary to provide adequate ventilation and cooling, which improves user comfort.

The air supply for SCBAs or for air supplied suits needs to be tested for quality. If the air comes from a compressor, it should be checked regularly. Similarly, the air from cylinders needs to be tested and the cylinders certified to ensure that the air meets breathing air standards. In the USA all breathing air must meet the requirements of ANSI-Z86.1 [87], which defines the composition and allowable contamination of several different grades of compressed air and prescribes methods of sampling and analysis. In Canada breathing air quality must meet requirements prescribed by the Canadian Standards Association [88].

Because of the hood noise resulting from the air flow, noise levels above 80 dBA (the limit for government contractors in the USA) make voice to voice communications very difficult without electronic equipment. Built-in communications systems, requiring cables in addition to the air hose, are often used. Battery powered two way radios are also popular. Many of these devices are of the voice or throat actuated type and leave the wearer's hands free. These communications systems are worn inside the hood of the suit. Communication devices of this type and others are available from many suppliers. Ontario Hydro uses special head-sets provided with



a noise cancelling dynamic microphone and amplifier that provide high intelligibility with little of the first spoken syllable missing [27]. Good communication capability should be regarded as an important safety feature. If workers cannot easily communicate while in the field, particularly in noisy work environments, they will tend to lift their protective hoods in order to talk.

In addition to providing adequate protection against tritium uptakes and acceptable communication capability, the suit must be designed so that it does not present conventional safety hazards. This means that it must provide good vision and freedom of movement, be non-flammable, not present slipping or tripping hazards and be quick and easy to take off in an emergency. Comfort is also very important to ensuring user acceptance. This requires an air supply with an adequate flow rate and appropriate temperature, and good fit and comfort under working conditions. The suit should also permit relatively quick and easy dressing and undressing.

Because of the danger of loss of breathing air and suffocation, and the difficulty of putting on and removing a supplied air suit unattended, the use of the suit often requires the presence of several persons in addition to those actually working in the suits. One person may have the responsibility of assuring that the breathing air supply is adequate and not interrupted. Another may be available to bring tools and equipment required by those who are suited and whose mobility is limited. Someone must be in communication with the person in the suit at all times and should be in visual contact as much of the time as possible. However, it should be noted that some suits, in particular those used by Ontario Hydro at its CANDU stations, are specifically designed for unaided dressing and undressing.

## **5. PROPERTIES AND COMPATIBILITY OF MATERIALS FOR SAFE PRACTICE IN TRITIUM CONTAINING SYSTEMS**

### **5.1. INTRODUCTION**

Apart from its radioactivity, tritium behaves physically and chemically very similarly to hydrogen and deuterium. The chemical forms of primary interest are tritiated water (HTO, DTO, T<sub>2</sub>O) and tritiated hydrogen gas (HT, DT, T<sub>2</sub>). Organic forms of tritium as well as metal tritides are also encountered. Knowledge of the behaviour of tritium and its interaction with materials is important in tritium safe handling and in the selection of tritium compatible materials and equipment. Physicochemical reactions are often catalysed or modified by the presence of ionizing  $\beta$  particles from tritium, by isotopic effects or by the presence of <sup>3</sup>He from tritium decay.

From a tritium safe handling standpoint, interest in tritium-materials interactions focuses on the following issues:

(a) Containment integrity

Tritium containment may be breached by mechanical failure due to corrosion, embrittlement or chemical reactions, by leakage due to chemical interaction of tritium with sealing materials, or by physical processes such as permeation.

(b) Contamination control

Tritium absorption in materials, out-gassing and migration by isotopic exchange and permeation through materials are important contamination control considerations.

(c) Tritiated compound production

Conversion of HT to the more hazardous HTO and production of tritiated ammonia, hydrocarbons and other organic materials may pose additional radiological hazards. Also, the products of tritium interactions with materials may require special consideration in air cleanup systems and waste handling.

Much of the following discussion on tritium interactions and compatibility with materials has been derived from Refs [89, 90].

TABLE V. RADIOLOGICAL PROPERTIES OF TRITIUM

Half-life	12.33 a
Maximum $\beta$ energy	18.5 keV
Average $\beta$ energy	5.7 keV
Mass	3.016 05 amu
Specific activity	355.9 TBq/g
Power density	0.324 W/g
Activity density of T <sub>2</sub> gas at 1.013 25 × 10 <sup>5</sup> Pa	95.8 GBq/m <sup>3</sup> (0°C)
	87.7 GBq/m <sup>3</sup> (25°C)

## 5.2. NUCLEAR AND RADIOLOGICAL PROPERTIES

Tritium decays by the emission of a weak  $\beta$  particle and the daughter product is  $^3\text{He}$ . Helium is a monatomic gas; therefore the decay of 1 mol of  $\text{T}_2$  yields 2 mol of helium, which causes a pressure buildup in sealed vessels containing  $\text{T}_2$ . The values of some radiological properties and derived parameters for tritium are given in Table V.

The  $\beta$  particle emitted from tritium decay interacts with matter by colliding with bound electrons in the surrounding material, producing electron-ion pairs, molecular excitations and free radicals and/or breaking chemical bonds. The range of  $\beta$  particles in aluminium, for  $\beta$  energies less than 2.5 MeV, is given by [91]:

$$R = 412 E^n$$

$$n = 1.265 - 0.094 \ln E$$

where

R is the range ( $\text{mg}/\text{cm}^2$ );

E is the  $\beta$  particle energy (MeV).

The range for a 5.7 keV  $\beta$  particle (average tritium  $\beta$  energy) is  $0.049 \text{ mg}/\text{cm}^2$  in Al, or 0.18 mm, since the density of Al is  $2700 \text{ mg}/\text{cm}^3$ . To derive the  $\beta$  particle range in a different material M, one must correct for the relative scattering probability per unit mass. Since scattering is from electrons, the higher the atom density per unit volume and electrons per atom, the shorter the range.

TABLE VI. RANGES OF TRITIUM  $\beta$  PARTICLES

Material	E ( $\beta$ ) (keV)	R'
$\text{T}_2$ gas, STP	5.7	0.26 cm
$\text{T}_2$ gas, STP	18.5	3.2 cm
Air, STP	5.7	0.036 cm
$\text{H}_2\text{O}$ , liquid	5.7	0.42 $\mu\text{m}$
$\text{H}_2\text{O}$ , liquid	18.5	5.2 $\mu\text{m}$
Stainless steel	5.7	0.06 $\mu\text{m}$

Therefore,

$$R'_M = R'_{Al} (\rho_{Al}/\rho_M) (Z_{Al}/Z_M) (N_M/N_{Al})$$

where

Z is the atomic (or molecular) number;

N is the atomic (or molecular) mass;

R' is the linear range.

Ranges of tritium  $\beta$  particles in various materials are given in Table VI.

### 5.3. CHEMICAL AND RADIOLYTIC REACTIONS

Tritium reacts chemically with a wide range of compounds and materials, either directly or through isotopic exchange. One special problem is the fact that the tritium  $\beta$  particle, with an average energy of 5.7 keV, carries more than 1000 times the energy required to break chemical bonds. Thus, many chemical interactions with tritium are radiolytically catalysed.

#### 5.3.1. Corrosion

Tritium reacts with halogen containing materials such as Teflon and Kel-F (which are used in valve packings, seats and gaskets), and with fluorinated pump oils, to produce tritiated corrosive halogenated acids. Teflon or Kel-F exposed to  $T_2$  produces the acid TF, which produces  $SiF_4$  gas in a glass system [92]. A mixture of  $T_2$ , moisture and Teflon in a stainless steel system maintained at high pressure (130 MPa) was found to develop catastrophic stress corrosion cracking of 0.76 mm thick steel in 16 h [93]. No failure occurred when  $D_2$  was substituted for  $T_2$  or when Teflon or moisture was removed.

The presence of  $T_2$  is incompatible with that of  $O_2$  in high level tritium handling, and  $O_2$  should be avoided as far as possible. Highly tritiated water is formed and can react aggressively with metals to produce severe corrosion due to peroxide evolution [94]. Tritiated water can also be produced through the interaction of  $T_2$  with oxygenated materials such as some organics and mineral oxides. Isotopic exchange between  $T_2$  and moisture is another mechanism of tritiated water production. The much higher radiotoxicity of HTO relative to that of HT and the corrosion potential of highly tritiated water underline the importance of eliminating  $O_2$  and moisture in  $T_2$  handling systems.

### 5.3.2. Degradation of structural properties

Tritium permeates and dissolves in materials and subsequently interacts with them chemically and radiolytically, resulting in degradation of structural properties that could lead to loss of tritium containment and contamination control problems. Examples of some of these interactions are:

- (a) Hydriding and embrittlement of metals;
- (b) Reactions with glasses, leading to the disruption of Si–O–Si bonds and the formation of Si–OT and Si–T bonds and hence the deterioration of mechanical properties;
- (c) Reactions with organic substances such as plastics, elastomers and oils, leading to hardening of materials (elastomers), changes in viscosity (oils) and the production of corrosive or volatile tritiated species.

### 5.3.3. Production of volatile tritiated compounds

Tritium reacts with carbon in steel and with thin oil films on the inner surface of steel piping and vessels to produce tritiated methane and other tritiated hydrocarbons. HT is converted to HTO through surface or radiolytically catalysed oxidation and through isotopic exchange with moisture in air or absorbed in materials. Tritium interactions with nitrogen and possibly nitrogenous materials can produce tritiated ammonia.

## 5.4. PHYSICAL PROPERTIES OF TRITIUM GAS

### 5.4.1. Solubility

Hydrogen (and HT) dissolves to some extent in almost all materials. From a safety standpoint, a higher solubility implies potentially higher tritium inventory in materials and contamination levels in wastes, as well as out-gassing problems. A higher concentration of tritium in materials will also lead to a potentially increased rate of chemical or radiolytic degradation. Hydrogen atoms or H<sub>2</sub> molecules and occasionally larger hydrogen bearing molecules dissolve in solids interstitially; that is, they diffuse into a structure and locate between atoms or molecular frameworks. As it dissolves, the hydrogen slightly disrupts the structural networks of the material and causes expansion.

For materials in which there is overall repulsion between hydrogen molecules or atoms and the material structure, the dissolution of hydrogen in the material is endothermic. Although solubility is low in such cases, it increases as the temperature increases. This is true for alumina [95], many metals [96] and organic polymers [97], and perhaps for glasses [95] at low temperatures.

For materials in which there is overall attraction between hydrogen and the material, the dissolution of hydrogen in the material is exothermic. The solubility is higher than for endothermic systems but decreases as the temperature increases. This holds true for hydrogen atoms in glasses at temperatures greater than 150°C [95], in silicon carbide [95] and in metals in which chemical hydride phases form [96].

#### 5.4.1.1. Solubility in polymers, glasses and ceramics

Hydrogen gas dissolves as molecular  $H_2$  in many materials. Dissolution of hydrogen in these materials is generally endothermic and is directly proportional to gaseous overpressure [98]. The materials in which molecular hydrogen dissolves include organic polymers and glasses at less than 150°C. Organic polymers generally have atomic scale voids between the twisted polymer chains and the hydrogen molecules seek these sites. Hydrogen dissolution in glasses is assumed to be similar [99]. Solubility of  $H_2$  in polymers is approximately  $0.1 \text{ cm}^3 \text{ (STP)/cm}^3$  at 300 K, while it is 10 to 100 times lower in glasses. Polymers are unique in that hydrogen bearing molecules larger than  $H_2$  can readily dissolve in them. Water and hydrocarbon vapours are known to enter and often swell polymeric networks [100]. These molecules can carry tritium.

At temperatures well above ambient, hydrogen gas reacts with and ‘dissolves’ as atoms in glasses [101, 102] and ceramics [103, 104]. In the equilibrium  $H_2 + M \rightarrow 2H \cdot M$ , solubility becomes proportional to the square root of pressure. In glasses at temperatures of 300°C or more, the equilibrium



becomes important. In ceramics such as alumina and silicon carbide at or above 500°C, hydrogen appears also to react and migrate as atoms:



The extent of molecular dissolution of  $H_2$  in ceramics (and crystalline materials in general) appears to be comparatively low.

#### 5.4.1.2. Solubility in metals

Hydrogen dissolves as atoms in metals [105–107]. The hydrogen apparently exists within non-hydriding metal lattices as protons, deuterons or tritons, with the electrons in a metal conduction band. Because some metals are endothermic hydrogen absorbers and others are exothermic, solubilities vary by approximately 10 to 15 orders of magnitude at room temperature. Figures 13 [108] and 14 [109] show

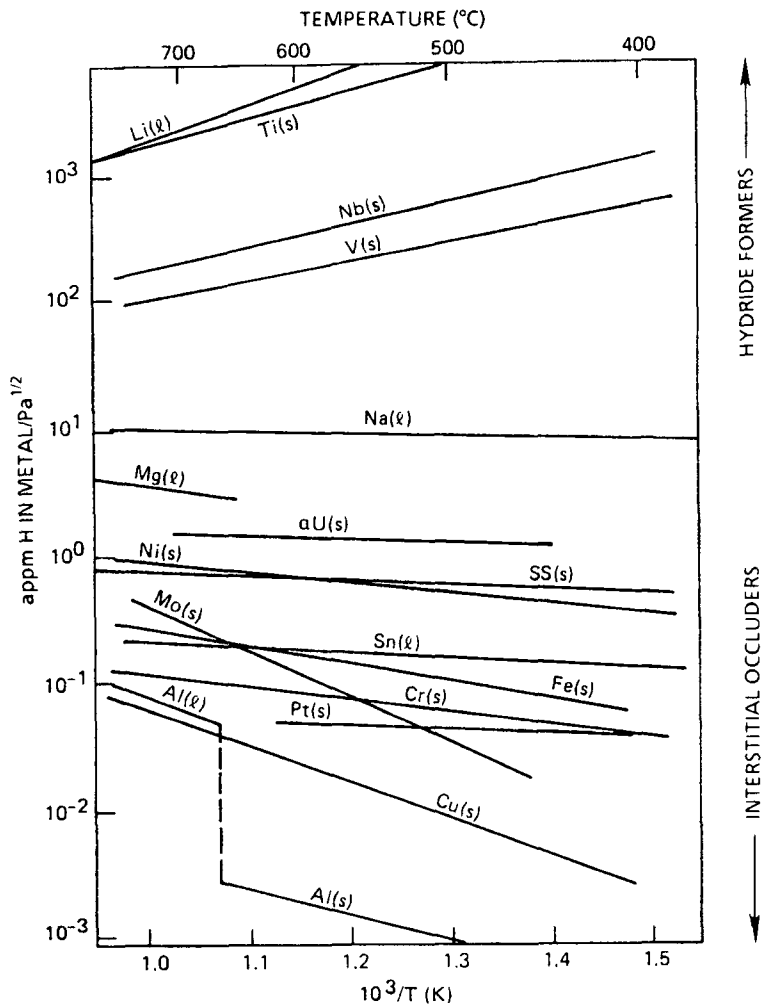


FIG. 13. Hydrogen solubility as a function of temperature for selected metals and alloys (s: solid; l: liquid).

hydrogen solubility as a function of temperature for various metals and refractories. Solubilities for endothermic absorbers give a negative slope (positive enthalpy) in the  $1/T$  plots, while exothermic systems have a positive slope (negative enthalpy). For various hydride phases, several of which have been used in tritium pumps and storage devices, plots of decomposition overpressure as a function of  $1/T$ , as shown in Fig. 15 [110], yield negative enthalpies or heats of formation.

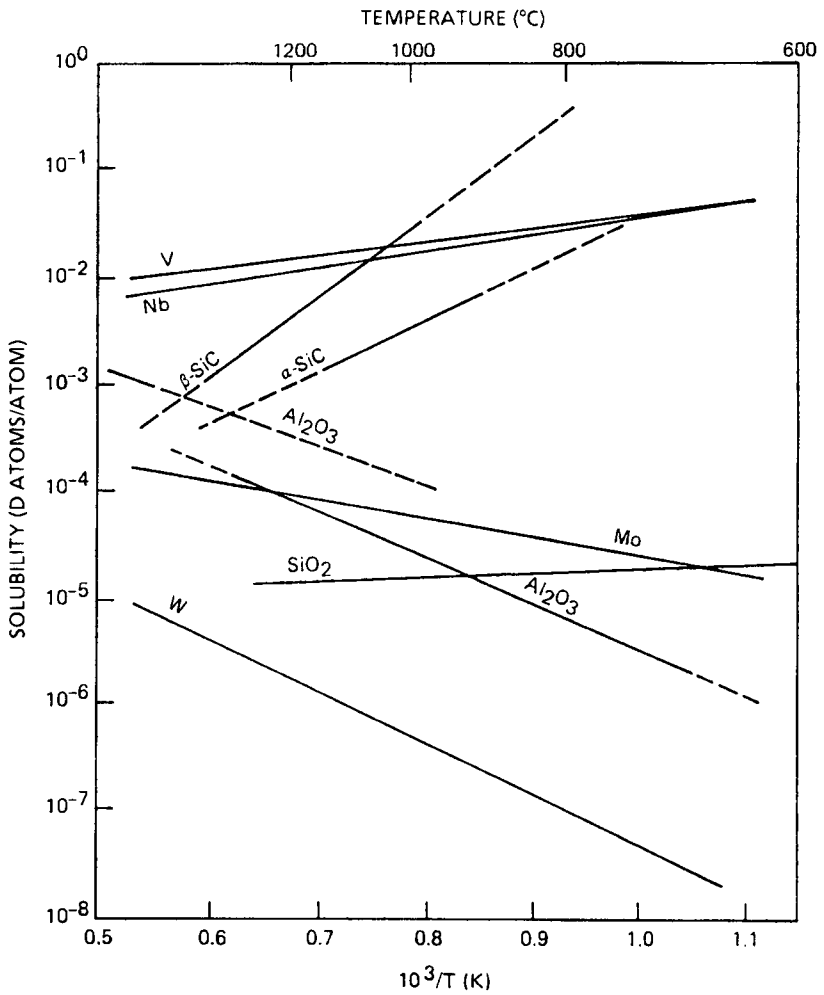


FIG. 14. Hydrogen solubility as a function of temperature for selected metals and refractories (broken lines: extrapolation).

#### 5.4.2. Diffusivity

The diffusion rate of tritium in material is given by Fick's law [111]:

$$J = -D \frac{dC}{dx}$$



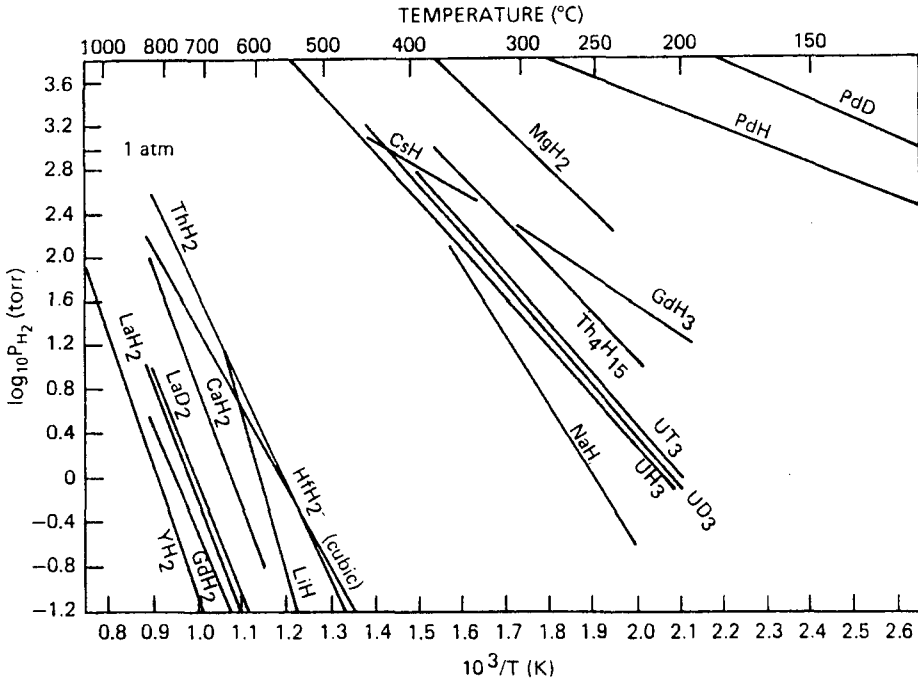


FIG. 15. Decomposition overpressure for various metal hydride phases as a function of temperature ( $1 \text{ torr} = 1.33 \times 10^2 \text{ Pa}$ ).

where

- J is the diffusion flux ( $\text{cm}^{-2} \cdot \text{s}^{-1}$ );
- $dC/dx$  is the concentration gradient;
- D is the diffusivity ( $\text{cm}^2 \cdot \text{s}^{-1}$ );
- C is the tritium concentration in material ( $\text{cm}^{-3}$ ).

The diffusivity of a material is a measure of its inherent ability to inhibit or control the movement of tritium through it. Values for common materials of interest vary over many orders of magnitude. Values of D as a function of  $1/T$  for hydrogen in various materials are shown in Fig. 16 [95]. Values for tritium are not expected to be greatly different.

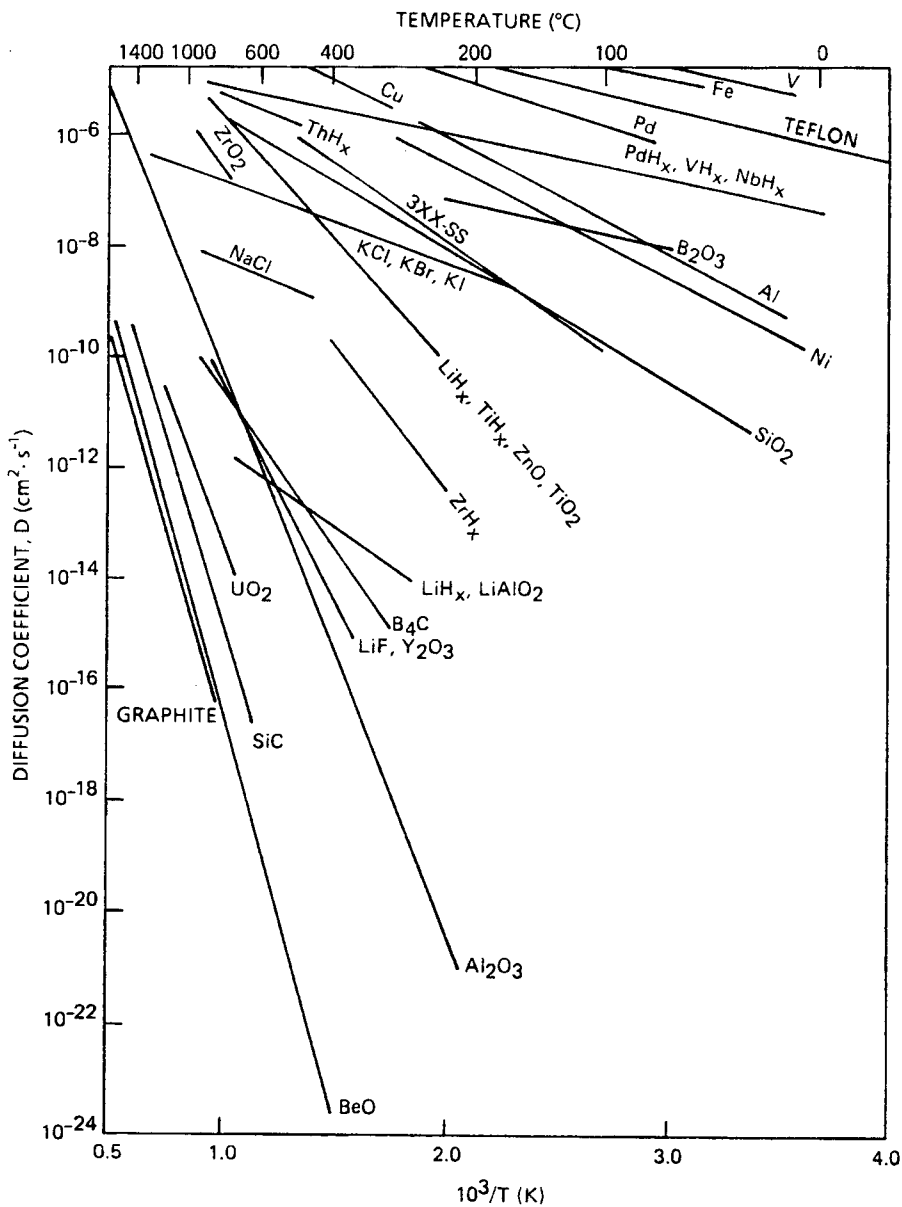


FIG. 16. Hydrogen diffusion coefficients in various metallic and non-metallic solids as a function of temperature. In several cases the plotted curves are averages from the work of several investigators.

### 5.4.3. Permeability

The equilibrium permeation rate of tritium through materials is:

$$J = \frac{D\Phi}{x} (P_1^n - P_2^n)$$

where

- J is the permeation flux ( $\text{cm}^{-2} \cdot \text{s}^{-1}$ );
- x is the thickness of material (cm);
- $P_1$  is the upstream partial pressure (Pa);
- $P_2$  is the downstream partial pressure (Pa);
- D is the diffusivity ( $\text{cm}^2 \cdot \text{s}^{-1}$ );
- $\Phi$  is the permeability.

In general the exponent n is 1 if the diffusing species is molecular hydrogen and 1/2 if the diffusing species is atomic hydrogen. It may be shown that  $\Phi = DS$ , where D is the diffusivity and S the solubility in the material. Permeability is a measure of how rapidly tritium will ultimately escape its containment. The permeation rate may be shown to be at equilibrium when  $Dt/x^2$  is approximately 1, where t is time in seconds.

The following materials are listed in order of increasing permeability: ceramics and graphite, silicates, non-hydriding metals, hydriding metals and polymers. Plots of  $\Phi(\text{H}_2)$  as a function of  $1/T$  for these materials are shown in Figs 17 [109] and 18 [110]. Permeability of many other hydrogen bearing molecules through polymers has been observed [99, 101]. For such molecules, permeability can be well in excess of that for  $\text{H}_2$  through a polymer, as seen in Tables VII and VIII [99] and from data given in Section 4. This must be considered when handling tritiated water or organic solvents.

Several factors influence the permeability of a metal, in particular surface conditions. Because the permeability of hydrogen through a metal oxide at a given temperature is usually orders of magnitude lower than it is through the metal, a thin surface oxide can markedly reduce permeability through the material [112].

Cold working stainless steels generally does not greatly affect the solubility, diffusivity or permeability of hydrogen [113]. An exception is 304L stainless steel, where induced martensitic transformation allows pathways of enhanced diffusivity [114]. In addition,  $\gamma$  irradiation of a permeating stainless steel sample does not affect permeability over several hours at dose levels of hundreds of rads [108]. However, tritium  $\beta$  irradiation at the surface of a permeating metal may over several months introduce defects into the surface oxide barrier layer, thereby increasing overall permeability.

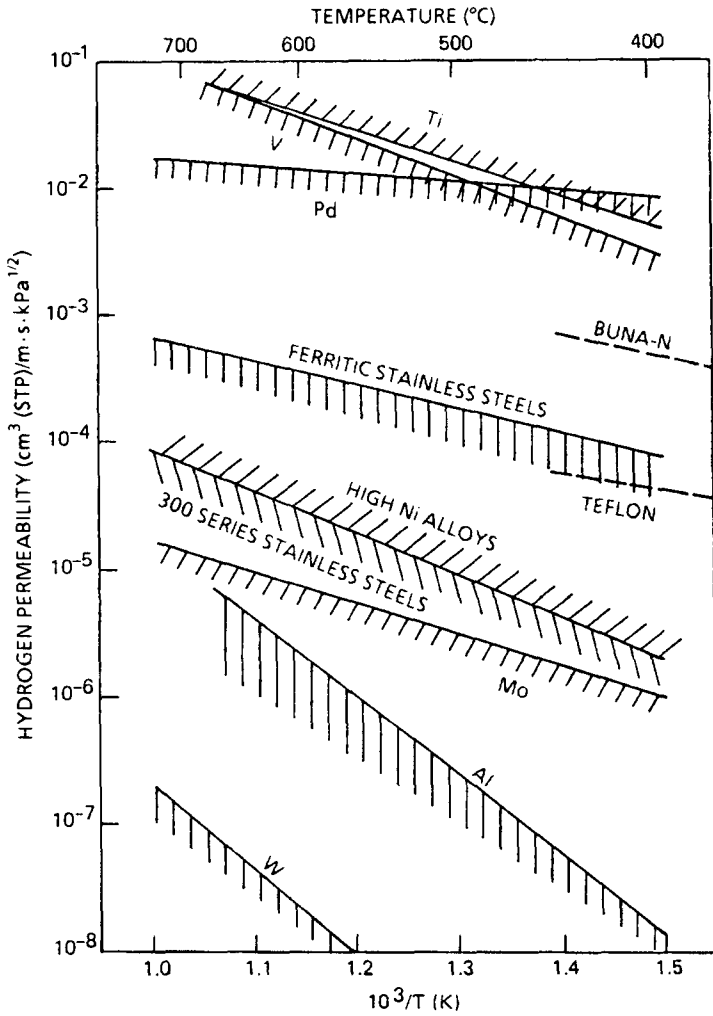


FIG. 17. Hydrogen permeability as a function of temperature for selected metals and alloys.

### 5.5. GUIDELINES ON TRITIUM COMPATIBLE MATERIALS

This section summarizes the key aspects of tritium-material interactions by class of material, from a safety standpoint, with recommendations on good practice in the selection and use of tritium compatible materials.

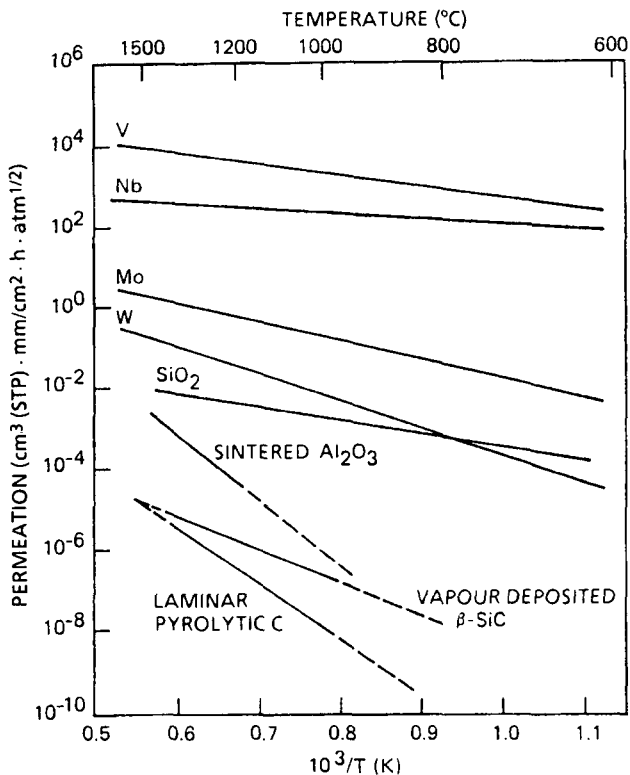


FIG. 18. Comparison of hydrogen permeation coefficients in refractory metals with those in  $\text{Al}_2\text{O}_3$ ,  $\text{SiC}$  and pyrolytic carbon at 1 atm ( $1.01325 \times 10^5$  Pa).

### 5.5.1. Metals

The mechanical integrity of non-hydrating metals in the presence of tritium is excellent because the electron bands carry away the energy of colliding  $\beta^-$  particles without disruption of metal structure or bonding [115]. These metals form the most common class of tritium containment structural materials. At high pressures of  $\text{T}_2$  gas, however, classical hydrogen embrittlement, as well as  $^3\text{He}$  embrittlement, can occur. For example, for 304L stainless steel samples exposed to 9 kpsi ( $62.1 \times 10^6$  Pa) of  $\text{T}_2$  at 423 K for 6 months and then aged 1.5 years, fracture toughness decreased by a factor of 6. Of this, a factor of 2 could be attributed to  $^3\text{He}$  alone [116]. Dramatic differences in ductility were observed between aged specimens charged with tritium and aged specimens charged with hydrogen [117].

TABLE VII. PERMEABILITY OF POLY(BUTADIENE-ACRYLONITRILE (35%)) RUBBER TO PENETRANTS AT 25-30°C

Penetrant	$P = \frac{(\text{cm}^3 \text{ (STP)}) (\text{mm} \times 10^7)}{(\text{cm}^2 \cdot \text{s}) (\text{cmHg})}$
Nitrogen	0.0042
Oxygen	0.016
Methane	0.032
Diisobutylene	0.045
Helium	0.084
Hydrogen	0.095
Carbon dioxide	0.13
Water	17.5
Methanol	246
Carbon tetrachloride	465
Ethyl acetate	3280
Benzene	6050
Methyl ethyl ketone	6100

Tritium charged specimens exhibited time dependent ductility losses due to the  $^3\text{He}$  pockets remaining in the lattice from tritium decay. Helium has been demonstrated to cause degradation in austenitic stainless steels at concentrations of the order of 200 to 700 appm.

The permeation rates of tritium through these metals are generally acceptable at temperatures below 100-300°C and for thicknesses of  $>0.1$  cm. For 304 stainless steel 0.3 cm thick with a 1000 cm<sup>2</sup> surface area exposed on one side to T<sub>2</sub> gas of 1 atm ( $1.013 \times 10^5$  Pa) pressure at 300 K, the steady state permeation rate is 5.9 MBq/d. However, at 800 K the permeation rate within a few hours is 20 TBq/d. The temperature dependence of permeation is often astounding.

Cross-contamination between metals and tritium occurs frequently enough to be troublesome. Oxide layers on metals often contain hydrogen and are further covered with a thin adsorbed carbonaceous film when originally grown in room air.

TABLE VIII. PERMEABILITY OF POLYMER FILMS TO WATER VAPOUR AT 25°C

Film	$P = \frac{(\text{cm}^3 \text{ (STP)}) (\text{mm} \times 10^8)}{(\text{cm}^2 \cdot \text{s}) (\text{cmHg})}$
Poly(vinylidene chloride) (saran)	0.3-1.0 <sup>a</sup>
Polytetrafluoroethylene (Teflon)	0.3
Butyl rubber	1.3
Polyethylene (density 0.960)	1.2
Polyethylene (density 0.938)	2.5
Polyethylene (density 0.922)	9.0
Polypropylene (density 0.907)	5.1
Poly(vinyl chloride)	6.1
Poly(vinyl chloride-vinyl acetate)	7.0
Poly(ethylene terephthalate) (Mylar)	13
Polystyrene	12
Polyacrylonitrile	13
Polybutadiene	47
Poly(styrene-butadiene)	9
Poly(butadiene-acrylonitrile (62%))	15
Polyisoprene (natural rubber)	30
Polyamide (nylon 66) (95% relative humidity)	68 <sup>b</sup>
Cellulose acetate	550
Cellulose acetate (15% dibutyl phthalate)	740
Ethyl cellulose, plasticized	1300
Poly(vinyl alcohol), $\rho = 2.3 \text{ cmHg}$	4200

<sup>a</sup> Dependent on grade.

<sup>b</sup> Dependent on relative humidity.

Upon later exposure to such a surface, tritium gas ( $T_2$ ) may become contaminated over hours or days with hundreds to thousands of ppm of protium (as HT) and methane (as  $CT_4$ ) as the surface layers are radiolysed, exchanged and chemically reduced [118–121]. The tritium exchanged into the oxide layer represents contamination of the material. Since diffusion of tritium in the bulk material is usually slow at room temperature, the extent of surface oxide contamination may greatly surpass the bulk contamination of a component. Minimization of cross-contamination may be accomplished by minimizing material surface areas, choosing an impermeable material with a thin or non-existent oxide layer and maintaining cleanliness.

Removal of tritium present in an oxide layer may be accomplished by acid dissolution of the oxide, or more gently by isotopic exchange with normal water [122–125] or activated  $H_2$  gas (i.e. plasma). Since diffusion of tritium dissolved in oxide or bulk material back to the surface of a material undergoing decontamination is often slow, exchange at an elevated temperature may be advantageous.

The recommended construction material for many tritium processing piping systems is austenitic stainless steel, typically 304 or 316, with a low carbon content. Copper is also recommended, especially at moderate temperatures and pressures. Pre-construction cleaning, using the methods described above, is important to minimize volatile tritiated product contamination.

For processing tritium at temperatures above 600 K, stainless steels may not be satisfactory because of the high permeability of iron/nickel alloys to tritium. Copper and aluminium have much lower permeabilities, but they also have reduced strength and creep resistance at high temperatures. One solution is to use a composite vessel with a liner of low permeability reinforced by a high strength outer shell. For temperatures above 800 K ceramic vessels will have to be considered. Alumina has very low permeability to hydrogen at high temperatures, but it is a very difficult material to fabricate into complex shapes.

## **5.5.2. Graphite, glasses and ceramics**

### *5.5.2.1. Graphite*

Because they generally have high surface areas, graphite samples adsorb large amounts of hydrogen gas ( $4 \times 10^{18}$  molecules/g for graphite pellets used in gas cooled reactors) [126]. Methane, protium and possibly water are generated from the  $\beta$  irradiation of the graphite surface [127]. The surface of the graphite will be contaminated with chemically bound tritium, and decontamination may be possible by baking at  $500^\circ\text{C}$  in the presence of a hydrogen exchange medium such as  $H_2$ ,  $H_2O$  or  $NH_3$ . Except for possible surface erosion, graphite will probably not be degraded mechanically over a period of several years, as bulk diffusion and solubility are extremely low.



### 5.5.2.2. Glasses

Various data suggest that HT in the presence of its chemically activating  $\beta$  irradiation energy could reduce silica bonding to  $-\text{Si}-\text{T}$  and  $-\text{Si}-\text{OT}$  species. At temperatures above  $300^\circ\text{C}$ ,  $\text{D}_2$  appears to reduce a silica network [101], and dissolved  $\text{D}_2$  in a  $\gamma$  irradiation field has the same effect [128]. The migration of  $\text{T}_2$  into glass structures could thus cause embrittlement and potential fracture over several months or years. Evidence suggests that activated hydration of glassy silica structures under  $\text{T}_2\text{O}$  exposure is also possible.

The permeability of hydrogen in silica glasses is one to two orders of magnitude greater than that for stainless steel over the temperature range  $0\text{--}200^\circ\text{C}$  (Figs 17, 18). This alone is not a major reason for rejecting silica containments of tritium. The exchange of tritium with naturally occurring hydroxyl groups in various glasses and on their surfaces is a source of protium contamination to tritium, perhaps 1% HT into a 1 atm ( $1.013 \times 10^5$  Pa)  $\text{T}_2$  supply within a 1 L glass container after 1 year [120]. Decontaminating a highly exposed glass of its bound tritium would require a significant number of water washes or hydrogen permeation flushes [129]. This effort is likely to be costly and is often not warranted by the value of the part undergoing decontamination. This is true for virtually all materials.

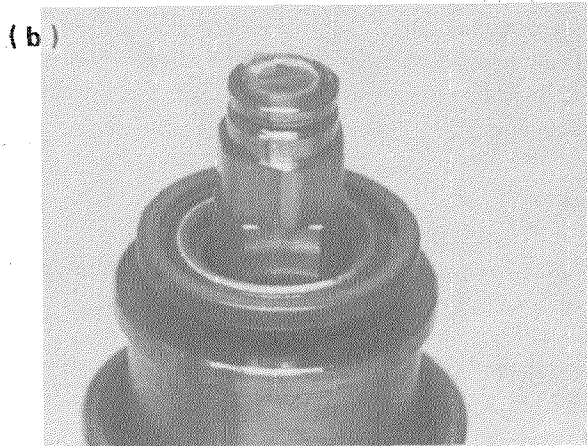
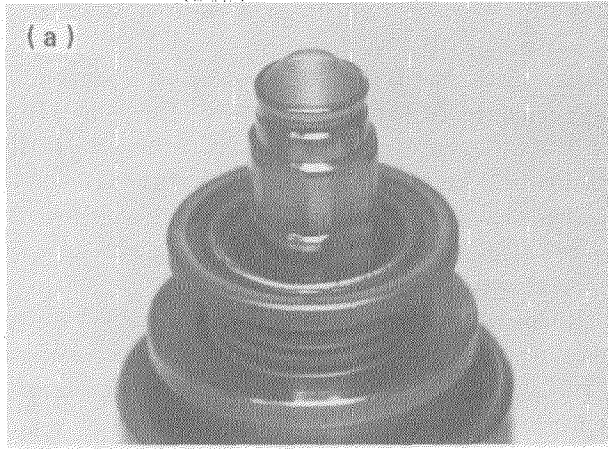
### 5.5.2.3. Ceramics

Because the hydrogen solubility, diffusivity and permeability are so much lower for ceramics than for glasses, ceramics undergo little or no bulk disruption from tritium. However, some mechanical degradation of regions near the surface is possible, with the depth of the area affected being a function of diffusivity and time. Oxygen release from  $\text{Al}_2\text{O}_3$  (sapphire) windows in the presence of liquid  $\text{T}_2\text{O}$  has been observed, although compatibility with  $\text{T}_2$  gas has been described as excellent [130]. The exchange of surface and near surface protium is likely, although mutual contamination of tritium and the ceramic should be less than that in the case of glasses. Tritium contaminated ceramics can probably be decontaminated by warm water or steam flushes or by etching in an acidic solution.

### 5.5.3. Plastics, elastomers and oils

Organics are readily permeated by tritium (gas or water) and are therefore subject to disruption of their bulk chemistry. There are few or no mechanisms for the rapid delocalization of  $\beta$  energy, and substantial mobility of organic chains occurs within polymer structures (particularly amorphous regions). Once formed, reactive organic intermediates can thus react with each other.

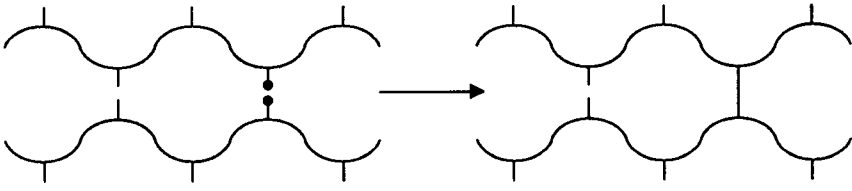
These effects are important when considering the design of tritium systems. Damage to components, such as gaskets, valve tips and O rings, must be carefully



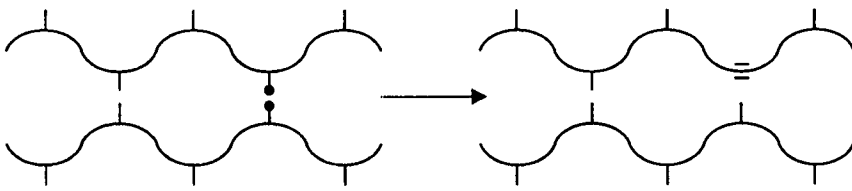
*FIG. 19. Kel-F stem tips on valves (a) before and (b) after exposure to 2 atm ( $2.026 \times 10^5$  Pa) of high purity  $T_2$  for two years. (b) shows the results of radiation damage. Fracture occurred at the stress point where the valve tip was closed against a stainless steel seat (courtesy of R.E. Ellefson, Mound Facility, USA).*

considered (see Fig. 19). Component failure during service can cause a serious release of tritium. Because elastomer seals often become embrittled, maintenance on nearby sections of piping may cause seals to develop leaks as a result of mechanical movement in the seal area.

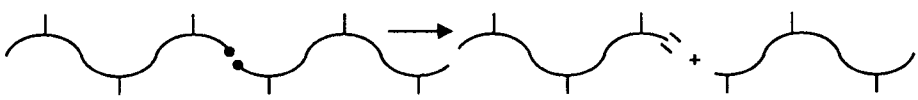
Figure 20 shows several polymer chain modifications that take place following activation by  $\beta$  radiation to ionic or excited species [131]. Cross-linking and degrada-



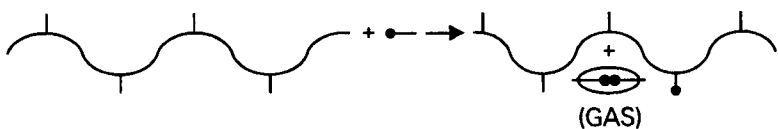
Cross-linking: the formation of chemical bonds between two polymeric chains



Unsaturation: the formation of double chemical bonds within the polymeric chain



Degradation: the fracturing of polymer molecules



Gas evolution: results in small molecules breaking away from the polymer

FIG. 20. Modifications to polymer chains that occur following activation by irradiation to excited states, ions or radical species.

tion are the most important processes affecting the mechanical properties of the polymer. These processes compete in a material, but those polymers which are most sterically hindered appear to preferentially degrade. Steric hindrance prevents neighbouring chains from linking and also imparts structural strains that are relieved upon chain scissioning. Cross-linking is indicated mechanically by an increase in tensile strength and a decrease in elongation, whereas degradation is evidenced by a decrease in tensile strength, an increase in elongation and softening of the polymer to a gummy consistency [132].

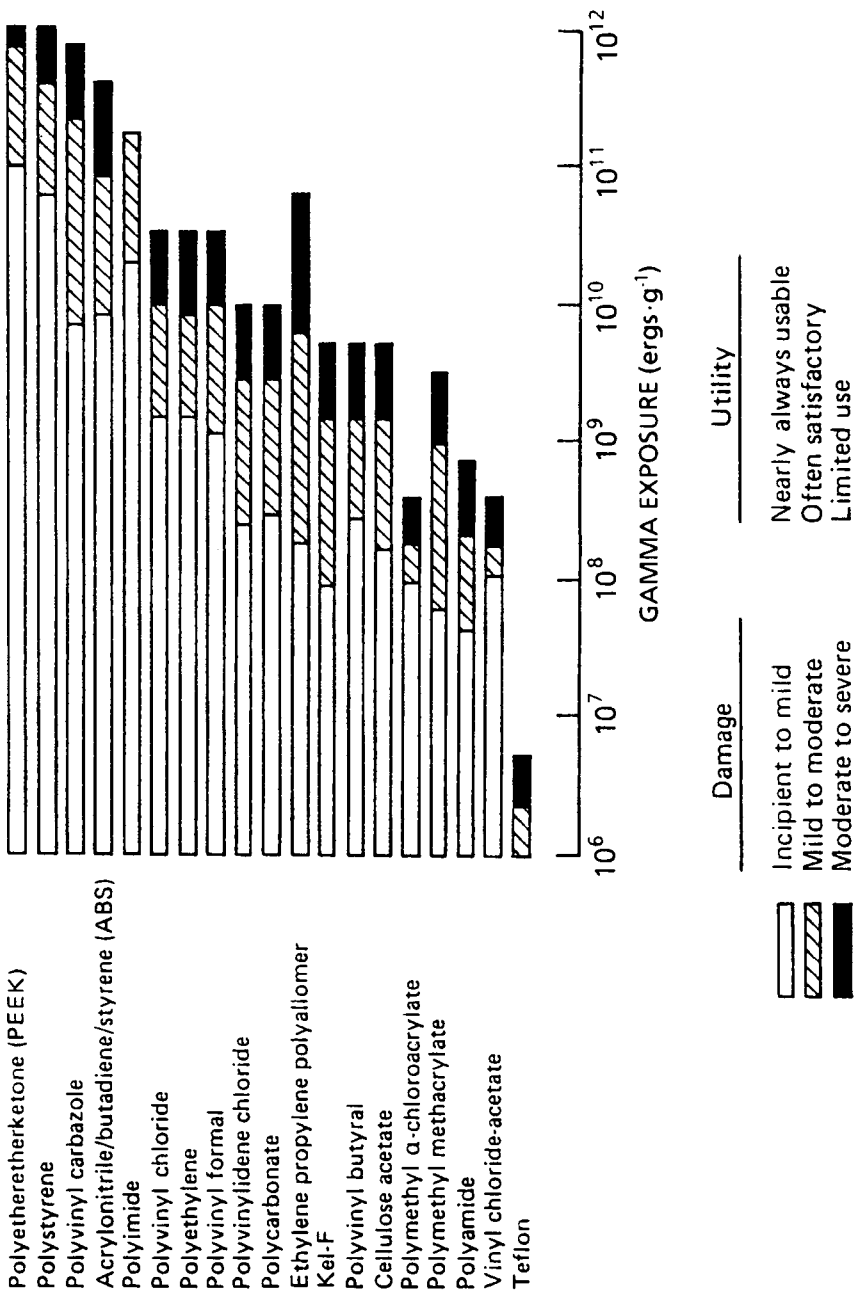


FIG. 21. Relative radiation resistance of thermoplastic resins ( $1 \text{ erg} = 10^{-7} \text{ J}$ ).

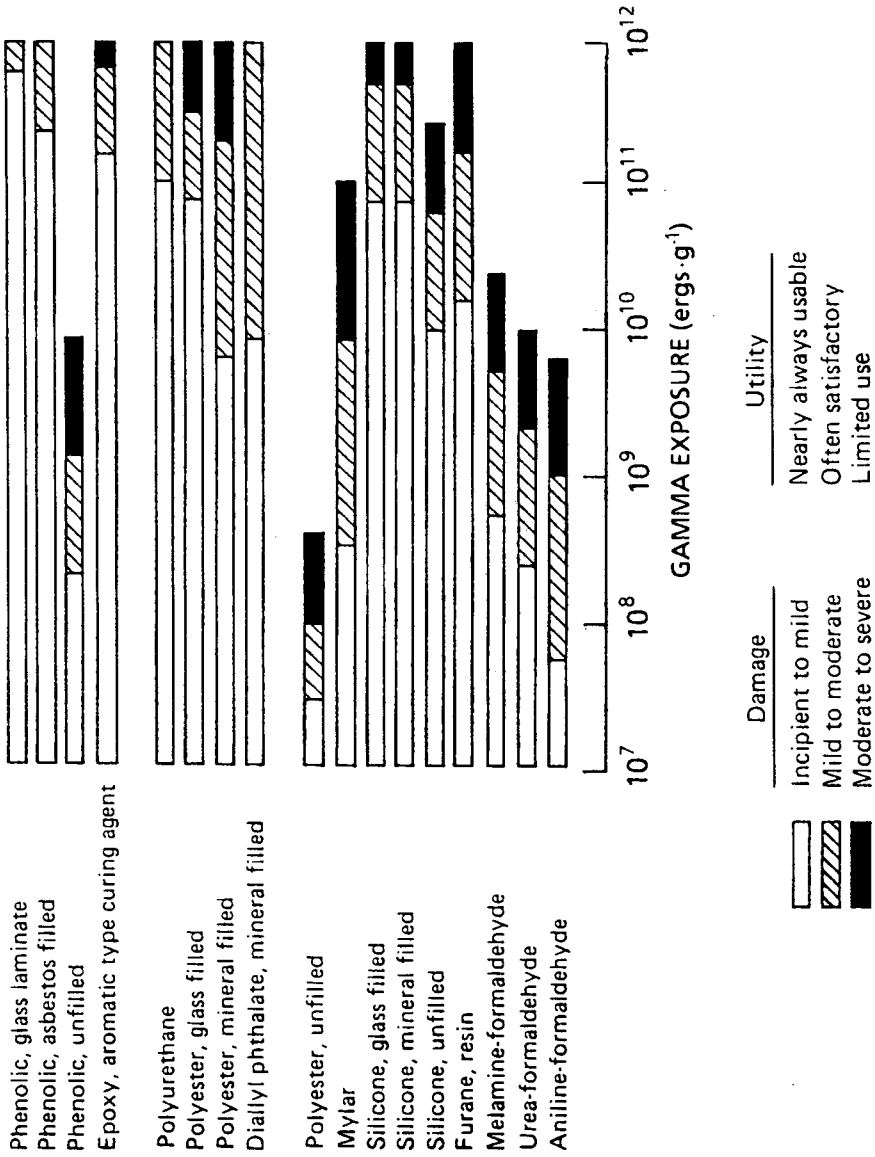


FIG. 22. Relative radiation resistance of thermosetting resins ( $1 \text{ erg} = 10^{-7} \text{ J}$ ).

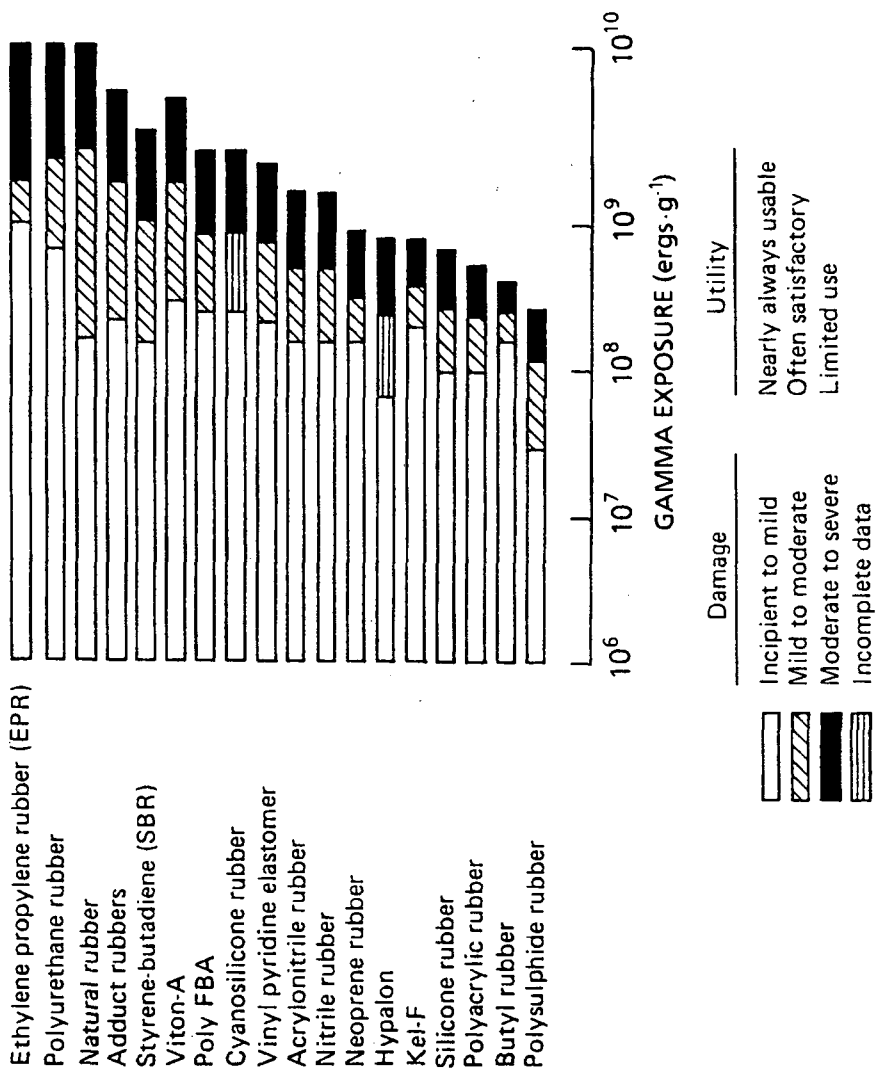


FIG. 23. Relative radiation resistance of elastomers (1 erg = 10<sup>-7</sup> J).

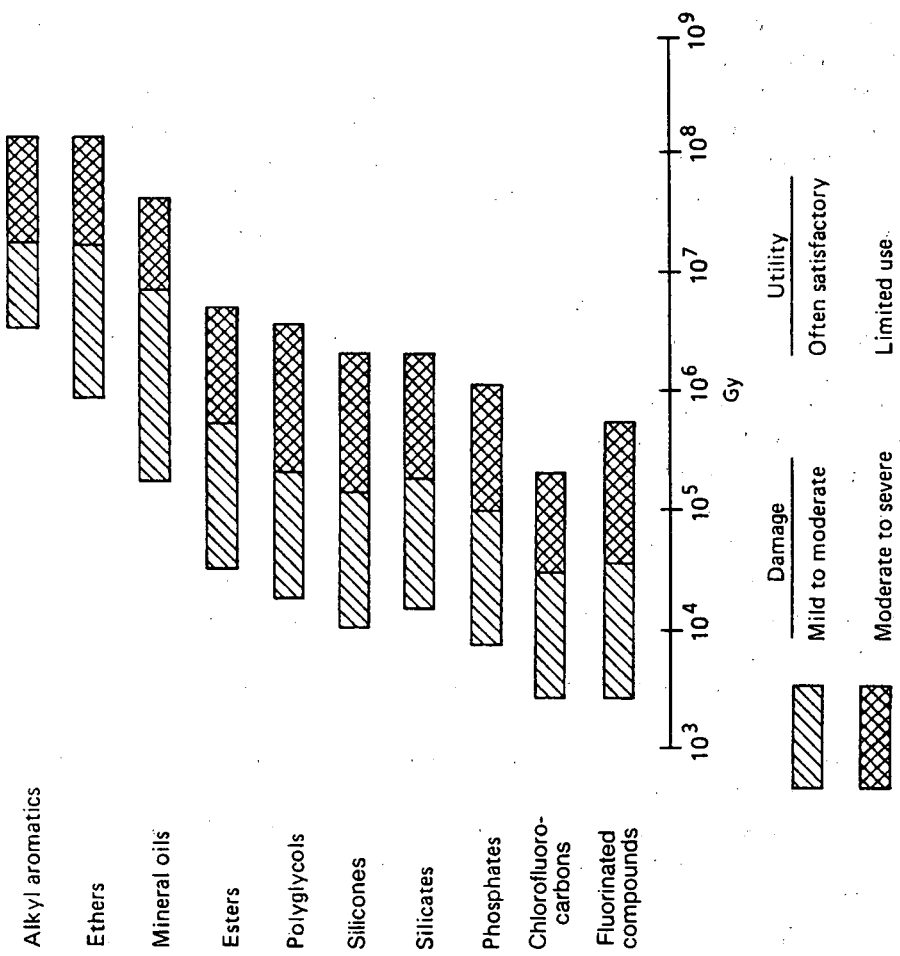


FIG. 24. Relative radiation resistance of base oils.

There are several factors that affect relative polymer stability. Energy-delocalizing aromatic structural groups increase polymer stability to distributing energies of excited states. Halogen atoms within polymers generate free radicals and thus promote radiation damage. Substituents on aromatic groups that extend the delocalized bonding network are further stabilizers. Saturated aliphatics are more radiation resistant than those that are unsaturated; isolated double bonds are readily excited to ions or radicals [131].

Organic compounds, in order of decreasing radiation resistance, are: aromatics, aliphatics, alcohols, amines, esters, ketones and acids. Figures 21-24 show the relative radiation resistance of plastics, thermosets, elastomers and oils to  $\gamma$  radiation [131]. Extension to  $\beta^-$  radiation is probably reasonable. In  $T_2$  gas, however, substantial differences in irradiation can occur between the polymer surface and the polymer bulk. This derives from the greater density of  $T_2$  (and the much greater range of the  $\beta^-$  in the  $T_2$  gas) outside the polymer compared with that inside the polymer bulk.

The relative increase in hardness of several elastomers after 87 days of exposure in 100 torr ( $13.3 \times 10^3$  Pa)  $T_2$  was investigated [92]. Hardening increased in the following order: natural  $\beta$ -butadiene  $\sim$  styrene  $\beta$ -chlorosulphonates  $<$  butadiene  $\sim$  acrylonitrile  $\beta$ -chloroprene  $<$  silicone (crumbled). This series follows quite well the relative stabilities as determined under  $\gamma$  radiation. Another experiment [133] noted the total compression set in several elastomers at 100 atm·d  $T_2$  exposure. Damage increased in the series ethylene propylene rubber  $<$  styrene-butadiene rubber  $<$  Viton  $<$  nitrile  $<$  urethane  $<$  silicone.

Surface versus bulk effects have been noted in some polymer- $T_2$  studies. Hardening of neoprene occurred throughout the bulk, while that of natural rubber primarily occurred at the surface (crack propagation) [92]. Total incorporation of tritium into a polyethylene powder was not a function of the amount of powder but of the exposed surface area [134].

Fluorinated materials such as Teflon and Kel-F used in valve packings, seats and gaskets should be avoided in tritium work involving  $T_2$  concentrations exceeding a few ppm. Apart from potential corrosion problems discussed in Section 5.3, degradation of these materials will lead to loss of sealing capability and hence loss of containment.

The solution is to use metal gaskets wherever possible and, where the resilience of an elastomer or the low friction of a plastic is necessary, to minimize the surface area exposed and use a non-fluorinated material such as polyimide. This appears to be acceptable for tritium service [135]. Valve stem tips made of Vespel, when used with sufficient sealing force, continue to seal for several years in  $T_2$  ( $\sim$ STP). When the material is employed with less sealing force, however, leaks have been observed across valve tips, possibly because of surface hardening. Polyimide gaskets under constant load are probably adequate for years. Studies [135]



showed that following exposure to T<sub>2</sub>, polymer hardness increased in the series: Vespel (polyimide) ~ Kalrez (perfluoro) < Viton (perfluoro).

Likewise, fluorinated pump oils are not recommended for T<sub>2</sub> service and certainly not for tritiated water vapour service, as TF evolution and corrosion may result.

Saturated hydrocarbon mineral oils (e.g. Duo-Seal) require frequent changes in T<sub>2</sub> service because of vapour pressure increases (off-gassing) and liquid viscosity increases. Silicone oils are rapidly polymerized or solidified. Polyphenyl ether oils last for years in similar service but are expensive and can absorb significant amounts of tritium.

## 6. MANAGEMENT OF TRITIATED WASTES

### 6.1. INTRODUCTION

The handling, packaging and storage of solid and liquid tritiated wastes are considered in this section. Gaseous wastes are either captured for subsequent tritium recovery, converted to a solid or liquid form, or released into the atmosphere via a stack. The final disposal of radioactive wastes has been discussed elsewhere [1, 2] and is beyond the scope of this report. It should be noted, however, that the storage and especially the packaging of radioactive wastes are chosen with ultimate disposal, usually by burial, in mind. More detailed information on the conditioning and disposal of tritiated waste from nuclear facilities can be found in Refs [1, 2].

### 6.2. ORIGIN OF WASTES

Tritium wastes originate from many sources. Some typical examples and their tritium concentrations are:

- Liquids from effluent capture systems (50 GBq/L to 500 TBq/L);
- Vacuum pump oil (<50 GBq/L to 5 TBq/L);
- Process residues (5-500 TBq/kg);
- Used or defective process equipment and piping (1 GBq/kg to 5 TBq/kg);
- Protective clothing (50 kBq/kg to 50 MBq/kg);
- Decontamination operations (50 kBq/kg to 500 MBq/kg);
- Process (hot) area office trash (<1 MBq/kg).

### 6.3. CLASSIFICATION OF WASTES

Wastes can be classified as low, intermediate or high activity wastes [136, 137]. Examples of wastes in each classification are given below along with examples of typical packaging for each classification.

Low specific activity wastes, such as soft trash, gloves, shoe covers, disposable clothing and paper, are generally compacted into simple containers such as 200 L carbon steel drums, or, in the case of bulky items, sealed in plywood boxes.

Intermediate level wastes, such as defective valves, fume hood gloves and trash and plastic suits, are generally placed within a sealed steel liner inside a 200 L drum. Larger items, such as process equipment, process piping and out-gassing items, may be placed in a welded steel box.

High activity wastes should be triply contained, for example: the primary container could be a welded steel container or a heavy-walled plastic drum; the secondary container could be a steel drum (100 L, for example) with the void filled with non-hardening asphalt or other material that minimizes the ingress of water and the escape of tritium; and the tertiary container is typically a 200 L steel drum.

### 6.4. DETERMINATION OF TRITIUM CONTENT

#### 6.4.1. Liquid waste

Generally the specific activity of liquid waste is relatively easy to measure by sampling and counting with a liquid scintillation counter.

#### 6.4.2. Solid waste

The distribution and the total amount of tritium are very difficult to determine in solid waste. In order to estimate the amount of tritium, information from the past history of the waste can be used. Experience in high activity tritium laboratories indicates that all parts of the process equipment that have been in direct contact with concentrated HT or HTO are highly contaminated. Such materials should be considered as intermediate or high activity waste.

The difficulties in determining the specific activity of solid wastes lead to the use of another parameter characteristic of contamination — the surface activity. This method is best suited to low or intermediate activity wastes. An estimate can be made on the basis of the amount of tritium removed by a smear (swipe). Experience in some tritium laboratories shows that, if the surface activity is less than 40 kBq/m<sup>2</sup> as measured by a smear, the waste may be considered as low level. Surface activities of less than 400 kBq/m<sup>2</sup> give rise to a room air contamination of less than

40 kBq/m<sup>3</sup> and, when leached by rain, to water contamination levels of less than 100 MBq/m<sup>3</sup>.

## 6.5. RELEASE OF TRITIUM FROM WASTES

Out-gassing measurements of tritiated waste may be made to determine the risk of air contamination associated with it. Several measurement methods are available or have been proposed [1, 136]. Out-gassing measurements are performed on the waste after packaging. However, it must be remembered that the out-gassing of a package at a given time is a function of prior handling and of temperature. It has been observed that a drum can have an out-gassing rate varying within a factor of 10 when the drum temperature varies between 0 and 20°C [2].

It is suggested that the measurements be performed when the package has not been disturbed and is at constant temperature.

## 6.6. PACKAGING AND CONDITIONING OF LIQUID WASTE

The purpose of this section is to examine packaging and conditioning of liquid waste for storage prior to disposal or tritium recovery.

For safety in conditioning, out-gassing caused by radiolysis should be taken into account for liquids containing more than 400 GBq/kg (10 Ci/kg). For these liquids a tight, strong container with provisions for pressure measurement and a pressure relief device can be used. Double or triple confinement is practised in some countries. However, in other countries it is permissible to ship liquid containing 400 GBq/kg of tritium in drums. A way to minimize radiolysis, out-gassing and the consequences of spills is to absorb aqueous liquids on sorbent materials and to store them. A second way is to store water as ice in a freezer.

Liquids with an activity less than 400 GBq/kg can be stored in tight containers with double or single confinement, depending on their specific activities and volume. It may be more desirable to sorb or solidify the liquid in an appropriate medium as part of the packaging process. In the sorption process, materials such as vermiculite, clay or diatomaceous earth can be used to sorb measured quantities of tritiated liquid. Even though these materials are being used, some disposal sites no longer accept absorbent materials and require a free standing solid for disposal.

Problems experienced with these absorbent materials include uneven distribution of the liquid on the material; desorption and movement of the liquid down through the package, causing a pool of liquid to collect at the bottom; and desorption and evaporation, followed by condensation at the surface of the package. Additional

problems may result from breaching of the waste container and subsequent release of the absorbent material into the surrounding medium. However, for small, laboratory scale quantities of tritium contaminated aqueous solutions with relatively low tritium concentrations, absorption onto such media may be an appropriate treatment prior to disposal.

Solidification of tritiated water with cement or other materials that form a monolith provides a waste package of high integrity that is more acceptable for disposal at many sites. Numerous commercially available solidification agents have been developed in an attempt to increase the load capacity for liquid radwaste. Load capacities ranging from 90 to 150 L of water per 200 L drum are typical. Agents that allow larger loading capacities are usually more expensive, resulting in little economic advantage. They also provide no better retention of the tritium. It should be noted that, regardless of the treatment provided prior to packaging, tritium is capable of migrating out of the waste package and into the surrounding medium to some degree. Barriers can significantly reduce this migration and will be discussed in the following paragraphs.

The package used most often for transportation and disposal of 'solidified' liquid wastes is a 200 L carbon steel drum similar to the United States Department of Transport Specification 17H or 17C 200 L steel drum. Low specific activity (LSA) waste, i.e. waste containing less than 10 MBq tritium per gram, can be packaged directly in the 200 L drums with no barriers other than thin wall polyethylene bags to ensure safe handling during packaging.

When concentrations exceed the LSA category and the total quantity of tritium in the waste package approaches 1 TBq, additional containment such as a rigid liner, either metal or plastic, is recommended. Tritium quantities that exceed 1 TBq per package may require additional barriers, such as high density rigid polyethylene liners. When the amount of tritium is in the range of tens of terabecquerels per package, asphalt barriers and multiple packagings, such as 110 L drums within 200 L drums, should be used to control tritium migration.

Some laboratories must package and dispose of rather large volumes of high activity tritium waste. Thirty litres of aqueous waste, containing up to  $10^{15}$  Bq, have been successfully packaged by solidifying the liquid in cement mix inside a 100 L high density polyethylene liner with a wall thickness of 1 mm. This liner is placed inside a 110 L 17H steel drum coated with an asphalt-bearing paint on the inner wall. A barrier of non-hardening asphalt is then placed over the top of the 100 L drum before the lid is placed on the larger drum. The 110 L package is then overpacked in a 200 L steel drum with an asphalt barrier at its top and bottom. Absorbent material is placed in the remaining annular space. Non-hardening asphalt has been demonstrated in tests to be an excellent barrier to tritium migration and is recommended for situations in which high activity tritium must be packaged for disposal.

## 6.7. PACKAGING OF SOLID WASTE

If the solid waste is compactible it is recommended that compaction be done as part of the packaging process to reduce the total number of waste containers to be discarded. Commercial compactors are available, although they may have to be modified to allow adequate containment of the contaminated air released during the operation.

Incineration may be a viable volume reduction technique for some combustible tritiated solid wastes. Most of the tritium in the off-gas will escape into the environment. With a wet off-gas handling system the possibility of capturing some of the tritium exists, but the tritium will appear in the liquid effluent.

Shredding or other volume reduction techniques can also be used to process solid waste contaminated with tritium. During the volume reduction process it is sometimes useful to apply a surface coating to the material in an attempt to fix any tritium near the surface while volume reduction is taking place. Paints or other spray coatings can be effective in retarding tritium release from surfaces.

Solid waste can be packaged in steel drums. Experience from tritium laboratories shows that out-gassing of drums will usually be less than 40 MBq/d per drum (1 mCi/d per drum) if the suggestions given in Section 6.3 for sorting at the source are followed. The drums can be stored in a ventilated room or building with air monitoring. The required ventilation rate will be a function of the number of drums in the room. The handling of such drums under normal conditions should not give rise to radiation exposure problems for workers.

It is recommended that all highly contaminated solid waste, such as process residues, be packaged in tight (or welded) containers for either interim storage before tritium recovery or disposal. Many of the packaging suggestions contained in the previous section are applicable here. The containers should be stored in a ventilated room with air monitoring.

## 7. SAFE HANDLING OF TRITIUM IN LABORATORIES

### 7.1. CLASSIFICATION OF TRITIUM LABORATORIES AND SUGGESTED SAFETY MEASURES

The IAEA Code of Practice on the Safe Handling of Radionuclides [138] limits the activity of unsealed sources that can be handled in various types of working place or laboratory, but no classification of tritium laboratories has been established. Such

TABLE IX. SUGGESTED SAFETY MEASURES FOR DIFFERENT CATEGORIES OF TRITIUM LABORATORIES HANDLING TRITIUM GAS

Safety measure	Category of laboratory		
	Low	Medium	High
Tertiary confinement	—	+	*
Double confinement	—	*	*
Zoned ventilation	+	*	*
Laboratory monitoring	*	*	*
Staff training	*	*	*
Exposure control	*	*	*
Bioassay and dose records	*	*	*
Stack monitoring	—	*	*
Recovery systems	—	+	*
Handling procedures	+	+	*
Emergency procedures	—	+	*
Environmental monitoring	—	+	*

**Note:** — not especially required; + useful; \* strongly suggested.

a classification is difficult because several parameters have to be taken into account, such as the inventory, form (gas, solid, liquid) and concentration of the tritium. Nevertheless it may be useful to suggest a classification. The following classification is based only on HT inventory, which is an adequate parameter in most cases. This suggested classification considers three categories of laboratory, according to the tritium mass inventory, M: low:  $< 0.01$  g; medium:  $0.01 \text{ g} \leq M \leq 1 \text{ g}$ ; high:  $> 1 \text{ g}$ .

Some general safety measures can be suggested for the three categories, but it is evident that each laboratory has to be considered individually, especially if more hazardous forms of tritium are used (such as HTO or pyrophoric compounds). Some of these measures are summarized in Table IX. This table provides general recommendations only. Application at a given site or facility must take into account site specific characteristics and local requirements.

## 7.2. SPECIAL PROBLEMS RELATED TO THE OPERATION AND MAINTENANCE OF TRITIUM SYSTEMS

Certain special issues and problems associated with tritium should be recognized in the design, operation and maintenance of tritium laboratories.

### 7.2.1. Mobility

The various gaseous and vapour forms of tritium are extremely mobile and may escape from containment through minute leaks. Gaseous HT permeates directly through structural materials, especially metals at high temperatures. Both HT and HTO readily dissolve in or adsorb on a wide range of metals and organic and inorganic materials, and out-gas from these materials. They undergo isotopic exchange with moisture and various organic materials. A major safety requirement in tritium handling laboratories is to control the escape of tritium by means of effective and efficient containment systems and contamination control practices.

### 7.2.2. Chemical forms

The principal chemical forms of tritium have very different relative radiotoxicities. The ICRP recommends DACs for HT and HTO that differ by a factor of 25 000. For other tritiated gases the radiotoxicity may be intermediate, with the DAC for the more water soluble gases closer to that for HTO. It is necessary to know the chemical forms being handled so that proper handling procedures can be used and the necessary precautions taken. All reasonable conditions and possibilities should be considered, such as maintenance and accidents, before deciding that a particular tritium compound will not be present. In practice it is difficult to exclude the possibility of HTO. Since HTO is so much more radiotoxic than HT, its presence should generally be assumed and proper precautions taken.

### 7.2.3. Chemical and radiolytic interactions

The chemical and radiolytic interactions of tritium with materials and the resulting compatibility issues have been discussed in Section 5. A variety of problems can arise if the designers and operators of tritium systems are not aware of these issues. For example, the permeation and diffusion of tritium through the walls of process equipment can lead to unexpected concentrations of tritium in working areas and releases to the environment. Radiation damage can lead to sudden failures and loss of containment and control. Also, hydrogen is flammable and suitable precautions should be taken to deal with the potential for fire and explosion.

#### **7.2.4. Effluent cleanup issues**

The escape to the environment of gaseous forms of tritium cannot be prevented by exhaust filters and must therefore be prevented by some other means, such as detritiation units for removing tritium from the atmosphere. However, if the captured tritium is in the form of HTO, which is much more radiotoxic than HT, careful consideration must be given to the desirability of atmosphere detritiation. If tritium is released as HT, most of it will be widely dispersed and rapidly diluted before oxidation occurs. If tritium is oxidized to HTO during the capture process, the retained HTO will be another potential release source which has to be carefully managed. Each tritium laboratory has to find the best compromise between the release of HT and the retention of the more hazardous HTO.

### **7.3. CONTAINMENT**

#### **7.3.1. Introduction**

There are several specific objectives to be considered when designing a tritium facility. These objectives include the protection of people and the environment, production of a desired product, safe disposal of waste materials and decontamination and decommissioning of the facility.

Certainly one of the most important objectives of tritium facility design is the protection of people and the environment. The people to be protected include operating personnel, plant personnel and the public. Protection of the environment includes limiting both liquid and gaseous releases. The best way to protect people and the environment from tritium is to keep the tritium in the process where it belongs and to capture any material that escapes.

To effectively control the mobility and contamination spread of tritium, a multiple barrier system is used. Typically tritium systems are doubly contained. The nature of the primary and secondary containments will vary with the specific type of work to be performed.

The primary containment is that which surrounds the tritium; it typically consists of the process equipment such as process vessel, piping, valves, pumps and measuring devices. The secondary containment is often provided by a glove box but in some cases may be a fume hood or enclosure exhausted to the building's active waste collection system or to the environment. In processes where solid material containing tritium must be handled, it is often not possible to provide double containment. In these situations the glove box becomes the primary containment envelope.

A tertiary containment or confinement system could consist of the room and building structure in which glove boxes, fume hoods, etc., are located and may have a collection and detritiation system for the air exhausted from the working areas. A



zoning and ventilation control system should also be implemented to minimize tritium contamination of the clean zones by air from the working areas.

When the effectiveness of a multiple containment system is evaluated, it is important to note whether an additional barrier will have a part in common with a primary barrier, which would represent a compromise.

### **7.3.2. Primary containment**

#### *7.3.2.1. Design principle*

One underlying principle of general design seems appropriate to the design of tritium facilities. This principle is that a simple, straightforward design will greatly enhance the probability of having a successfully operating and safe tritium facility. For example, using manually operated valves ordinarily results in simpler designs, a more leaktight system, less maintenance and lower equipment costs. Although it is not always possible to use simple systems, they are preferable when there is a choice.

#### *7.3.2.2. Piping systems*

The process piping system usually is the primary containment system for tritium. Therefore, it demands careful attention during design and installation. The ideal system is an all-metal system compatible with hydrogen and containing nothing that is susceptible to hydrogen embrittlement or radiation damage. The recommended construction material for most systems is austenitic stainless steel, typically 304 or 316, with a low carbon content. However, copper and aluminium have also been used successfully for low pressure systems. Copper and aluminium have lower permeability to tritium than does steel but have reduced strength and creep resistance at higher temperatures. It is more difficult to weld aluminium reliably, and joining it to components made of other materials can be difficult.

Pre-construction cleaning or treating of the interior of process equipment is important for maintaining product purity. Tubing used in process systems should be cleaned or treated by a method that will achieve a surface reduction (reduced roughness) inside the tubing of at least 90%. Piping and tubing should be welded together to eliminate any leakage at the joints. For valves, pumps and other major equipment that requires disassembly of joints, engineered couplings with demountable metal gasket seals are recommended. Pipe threaded joints should be avoided.

The use of all-metal components is highly recommended so that no oils, plastics, elastomers, organics or fluorocarbons come into contact with tritium. These all-metal components are available commercially. An all-welded system will provide the highest degree of leaktightness. In practice, because valves, transducers and other

system components have limited life, it may be advisable to use demountable components and standard fittings, which can be replaced without cutting, welding or brazing tritium contaminated tubing. Experience has shown that a properly designed and assembled tritium system containing hundreds of demountable components can have leaktightness rivalling that of an all-welded system [139]. Tritium compatible components and equipment are discussed in Section 5.

#### *7.3.2.3. Valves and fittings*

Packless valves of the metal bellows or diaphragm type are recommended for tritium service. They may be remotely operated with pneumatic actuators, which are convenient for glove box operations. Manually operated valves have the advantage of simplicity but should be used with torque limiting handles whenever possible. The valve seat or tip should be made of metal or of a non-fluorinated, radiation resistant material such as polyimide. High vacuum fittings with demountable metal gasket seals are commercially available. For tritium work in glove boxes, types of fittings that do not require axial clearance for disassembly are particularly convenient, as components mounted with them can be removed without disturbing the connecting tubing. This type of fitting is used for tubing sizes up to 25 mm. For larger tubings, types of flanges and fittings such as those shown in Fig. 25 may be used. Other tritium compatible fittings are described in Ref. [89].

A double-valve purge system (Fig. 26) is recommended for connecting system components in a way that minimizes tritium contamination during the replacement of components. The vacuum system pumps out all contaminated gases between the valves and can also be used to introduce clean gas into this volume. The joint can then be opened with minimum contamination.

#### *7.3.2.4. Pressure protection*

Systems that handle tritium and those in which integrity must be ensured should be conservatively designed and helium leak tested. The helium leak tests should be performed at a pressure that corresponds to the maximum allowable operating pressure of the system. However, the applicable codes in particular jurisdictions may define the pressure to be used in specific tests.

Another consideration should be the need to helium leak test the system with rupture discs in place and also to leak test various components within the system that have different pressure ratings. All tritium systems should be designed with some form of overpressure protection, such as the use of rupture discs. These discs are constructed to be leaktight under normal operating conditions, whereas relief valves have been found unacceptable because tritium leaks occur through the valve seat seals.

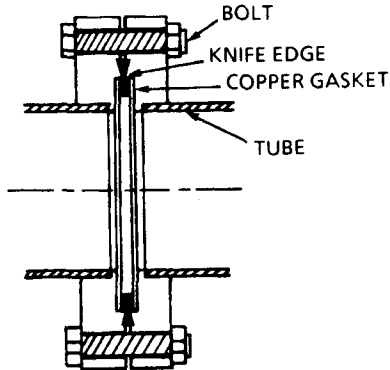


FIG. 25. Vacuum flanges and fittings.

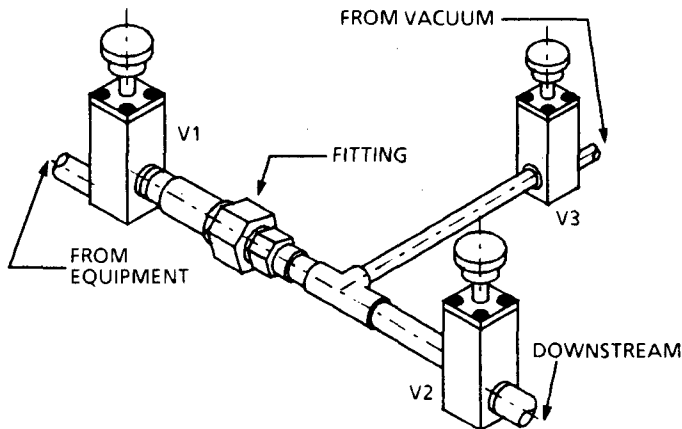


FIG. 26. Maintenance valve arrangement for replacing contaminated tritium system components.

Rupture discs may be vented to a secondary evacuated catch tank that is also rupture disc protected and is designed to accept the maximum credible volume release. This will enable lost tritium from the primary vessel to be recovered. The disc of this catch tank may be vented to the normal tritium capture system.

#### 7.3.2.5. Pumps

Ideally, tritium handling pumps should be dry, hermetically sealed and pack-less, with no oil lubricant, elastomers or fluorocarbons. Many different means of pumping tritium are used. Metal hydride beds can be used both as pumps and as

storage units. The ideal pump would have positive displacement, reliability, convenience and the ability to pump from a few dPa (mtorr) to over 200 000 kPa. However, no available pump has all these characteristics. Commonly used pumps include rotary vane types with magnetic drives (which eliminate moving seals), lubricated with low vapour pressure polyphenyl ether oil; metal diaphragm pumps; metal bellows pumps; scroll pumps; cryogenic compression pumps; and a combination of the metal bellows and scroll pumps. A metal bellows compressor has been modified to eliminate fluorocarbons from the pump chamber and used as a tritium gas transfer pump [139]. The principle of operation of the metal bellows pump is illustrated in Fig. 27. For high vacuum performance, the metal bellows pump may be supplemented by another bellows sealed pump with an orbital scroll geometry (Fig. 28). Other mechanical pumps with hermetically sealed drives include the magnetic bearing turbomolecular pumps and rotary vane pumps with magnetic drives (Fig. 29). Experience has shown that polyphenyl ether is satisfactory as a lubricant in such pumps.

For general purpose rough vacuum support for tritium systems requiring a capacity of less than 0.5 m<sup>3</sup>/min, rotary vane vacuum pumps are often recommended. The vacuum pump can exhaust to the atmosphere or to a tritium capture system. For systems in which the vacuum pump is connected directly to a tritium vessel, a pressure switch and automatic valve that will close if a set pressure is

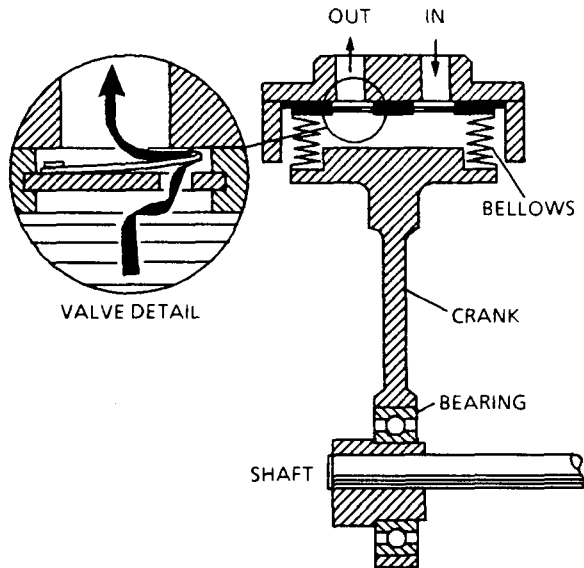


FIG. 27. Metal bellows vacuum pump (courtesy of Metal Bellows Corp., USA).

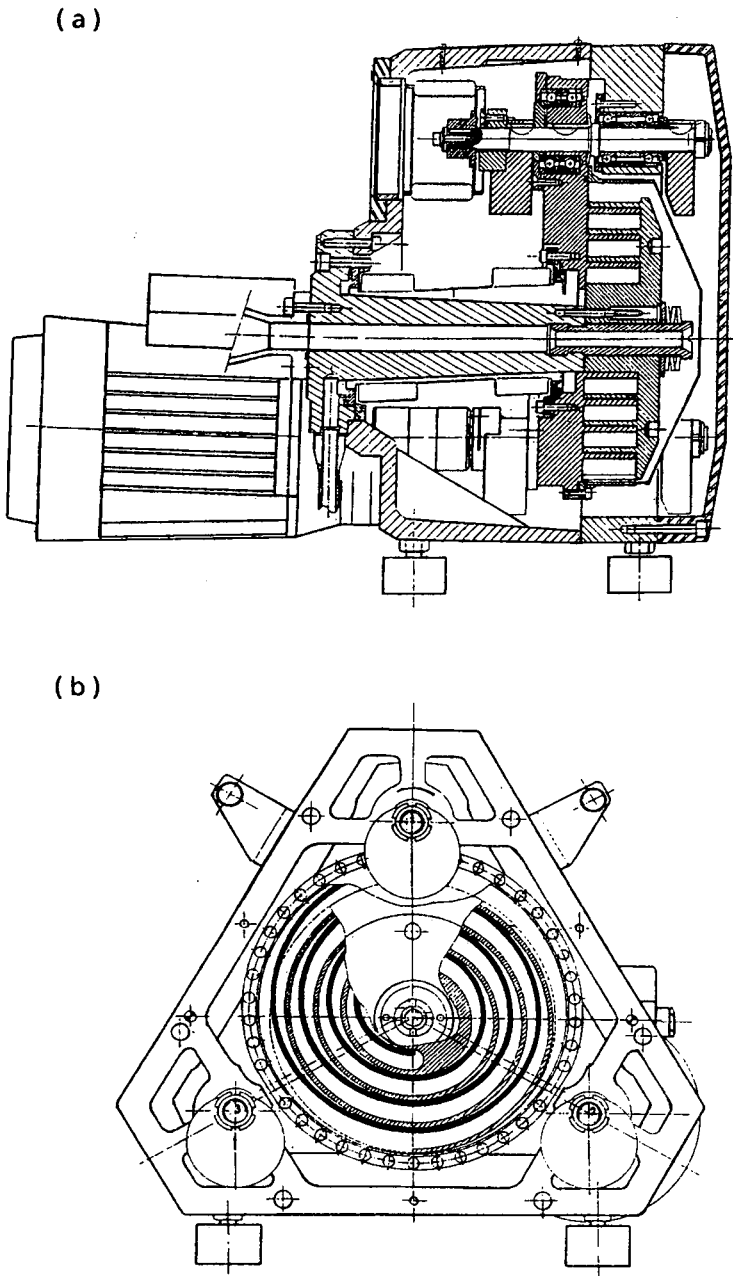


FIG. 28. Cross-section of pump showing (a) the flow path and (b) the vane assembly (courtesy of Normetex America, Inc., USA).

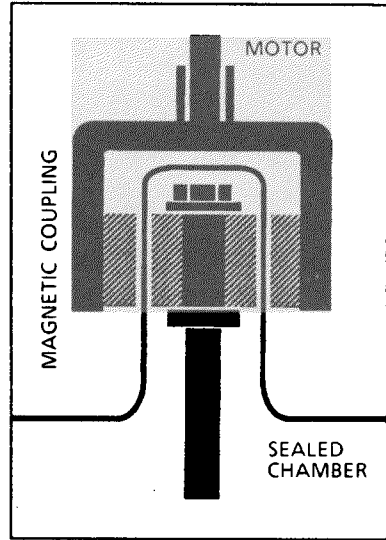
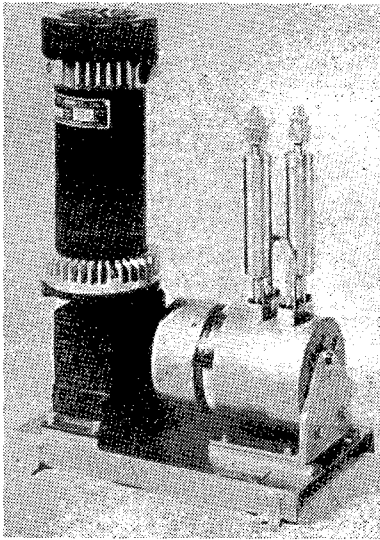


FIG. 29. Rotary vane pump with magnetic drive. Hatched area shows magnetic coupling (courtesy of Nova Scotia Research Foundation Corp., Canada).

exceeded should be used on the pump inlet. An alarm and a manual override for handling any expected higher pressures are usually included. The purpose of this is to protect the pump and safeguard against a major release of tritium in the event of a sudden rise in pressure due to inleakage. Periodic changes of pump oil can be accomplished by feeding fresh oil to the vacuum pumps from outside the glove box. The used oil then goes to a receiver tank and finally into a piping system that holds spent oil. Direct transfer of oil from the spent oil tank inside the glove box to a waste drum outside the glove box has proven to be an efficient and safe way to dispose of waste oil.

### 7.3.3. Secondary containment

#### 7.3.3.1. Introduction

There are several ways to provide secondary protection for tritium systems. The two most common methods are to enclose the primary system in a fume hood or in a glove box. Either method can be designed to protect the operating personnel. The difference between the two methods lies in their ability to prevent tritium from reaching the environment. In a typical fume hood any tritium released from the

primary containment will be released to the environment. If the amount of tritium being handled is small enough that its release to the environment is acceptable, fume hood containment may be appropriate. Each site must decide what constitutes an acceptable tritium release on the basis of its individual features. In general, glove boxes or other true secondary containment systems (systems that can completely contain all tritium that escapes from the primary containment system) are efficient at protecting both the operating personnel and the environment.

During the design of the secondary containment it is important to prevent the possibility that both primary and secondary containment systems will fail simultaneously. The most common error is constructing a secondary system that cannot withstand the pressure generated by a primary containment failure. A common example is a glove box that has insufficient passive relief capacity to deal with a large leak from the primary system.

The form of the tritium (elemental or oxide) must also be considered. The oxide (HTO) is four orders of magnitude more hazardous than the elemental form; the design of the secondary containment should reflect this. In addition, HTO permeates through gloves 10 to 100 times faster than HT. To counteract these properties of the oxide, it may be necessary to dry the glove box atmosphere to reduce the HTO concentration. It may also be necessary to minimize operator in-glove time or use double gloves on the glove boxes. When some or all of these precautions are taken, high concentrations of tritiated water can be handled safely in the glove boxes.

#### 7.3.3.2. *Glove boxes*

Glove boxes for handling large quantities of tritium are usually high integrity enclosures fabricated from welded stainless steel (type 304 is a popular choice) or aluminium with an appropriate number of windows and glove ports to accommodate the work to be done in the box. The metal is usually 0.32–0.48 cm thick. The corners and edges are preferably rounded and the stainless steel polished to give a smooth surface in order to facilitate cleaning. The window material can be laminated safety plate glass, usually 9–12 mm thick, polyacetate (Lucite, Plexiglas, Altuglass), or polycarbonate (Lexan or Makrolan). The windows should be sealed to the enclosure with soft rubber (silicone has been used successfully) gaskets to ensure a good seal and therefore a low leak rate.

Gloves for the box should be selected for the specific application while keeping in mind the need for ruggedness and resistance to permeability by hydrogen and water.

The ultimate requirement is for the box to be leaktight. Many boxes have been constructed that are helium leaktight, with probe detection levels less than  $10^{-6}$  cm<sup>3</sup> (STP)/s at a pressure differential of 15–20 cm of water.

Once the decision has been made that containment is required and that glove boxes will be used, it must then be decided whether to use an air atmosphere or some

inert gas. Air atmospheres are cheaper and easier to maintain than inert atmospheres but they have some disadvantages. For example, following a release of tritium gas into the glove box an explosive mixture may be formed. If protium or deuterium gas is also contained in the system, the hazard is increased accordingly. The oxygen in the air may also result in higher concentrations of HTO in the box. Also, unless the air supply is dried, the moisture in the glove box atmosphere can become tritiated, diffuse through the gloves and cause exposure of the workers. Some of the tritiated water can also sorb onto the surfaces inside the glove box, leading to a continuing source of HTO and also increasing the difficulty of subsequent decontamination efforts. The moisture level in the glove box should be kept as low as practicable.

An inert atmosphere provides protection against fire and explosion and reduces the problems associated with HTO formed inside the glove box. From a safe handling viewpoint, dry nitrogen is a satisfactory choice in many cases and is less expensive than other alternatives. However, the particular materials being handled, or the process used in the glove box, may require the use of a specific inert gas and may also establish the tolerable levels of trace contaminants. The oxygen level in an inert atmosphere box should be maintained well below the explosion limit and the moisture level should be kept very low.

The use of an inert gas as the glove box atmosphere also makes it possible to recover tritium that leaks from the primary containment without the necessity of first converting it to the more toxic tritium oxide. This can be done by employing suitable getters in a variety of ways. The tritium is retained in the getter and can be either recovered, stored or sent for disposal.

Glove box atmospheres are operated at pressures either slightly negative, slightly positive or neutral with respect to the room atmosphere pressure. The choice is often dictated by the type of glove box atmosphere cleanup system employed, if any. A negative pressure atmosphere is usual where the gas passes once through the box, either to a ventilation system or to a control cleanup system. A positive pressure may be required to minimize the entry of air into the box in cases where the oxygen and moisture levels must be kept to a few parts per million. Glove box atmospheres may be static and purged on signal when a certain tritium concentration is reached, or continuously purged at a low rate, which is increased following a tritium signal, and directed to a detritiation system.

Equipment contained in a glove box may have to be moved in and out of the box. To prevent the release of tritium to the room air, this should be done through a transfer port, chamber or 'pass box', which can be evacuated and backfilled with clean gas. Provisions to monitor the tritium contamination in the transfer chamber are desirable. Doors to the pass box should meet the same leaktightness requirement as the glove box. The doors should exit into a fume hood that will act as a contamination control device for items exiting the glove box. Figure 30 illustrates an arrangement for transfer operations between the glove box and a fume hood. In laboratories where no routine material movement is required, a bag-out or trash/maintenance port



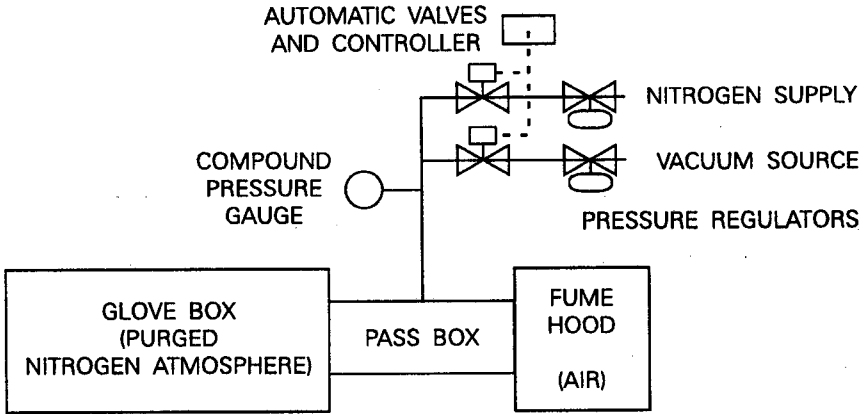


FIG. 30. Glove box-pass box-fume hood exit.

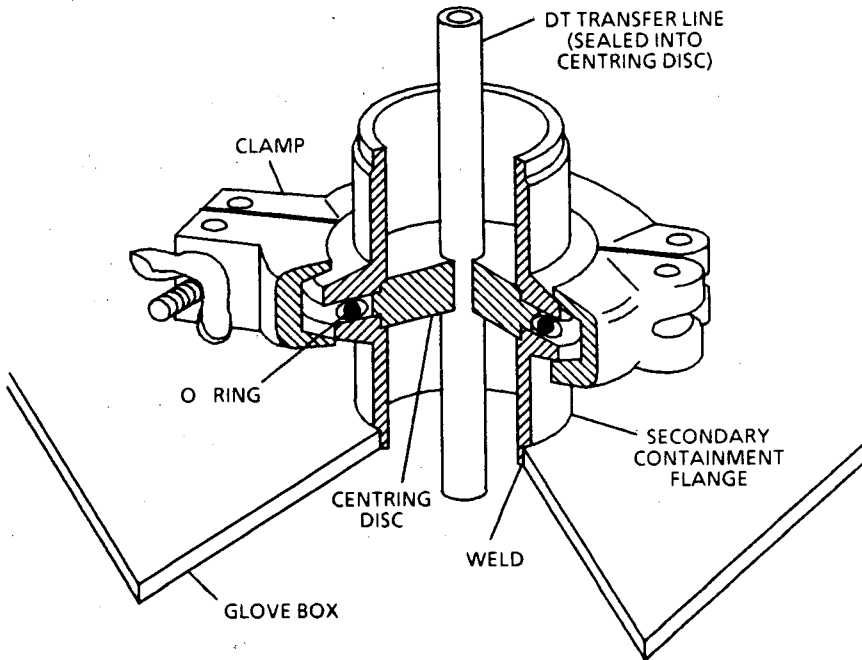


FIG. 31. Glove box penetration for tritium piping, complete with secondary containment jacket.

is usually acceptable. Materials sealed in a plastic bag can be moved into or out of the glove box through the port.

Where tritium lines have to be run from one glove box to another, they should run in a secondary containment jacket concentric with the tritium line. This jacket needs to be gastight and should be filled with the glove box atmosphere. The type of vacuum fitting illustrated in Fig. 31 provides a convenient means of assembling this conduit [89]. The same flange system, slightly modified, may also be used to provide leaktight feedthroughs of all types (pneumatic, wiring, instrumentation, etc.) into the glove box, as described in Ref. [139].

### 7.3.3.3. *Cleanup of glove box atmospheres*

No matter how good the primary tritium containment system, there will be leaks or releases to the secondary containment. Therefore, at times it will be necessary to remove tritium from the space between the primary system and the secondary containment. This can be accomplished by moving gas ( $N_2$ , Ar, He or air) through the secondary space to a tritium removal system. This can be done with a once-through purge, a closed recirculation loop or some combination of the two. The process most commonly used for such a system first oxidizes any hydrogen in the gas, including tritium, to water and then dries the gas. The tritiated water is collected for further processing or disposal. The detritiated dry gas is recirculated into the secondary system or released into the atmosphere. A third option, which must be carefully designed, is to purge the secondary containment directly to the stack when the tritium concentration is low, or directly to the tritium removal system when the concentration is high. This method requires a fast, reliable tritium monitor and a primary process that releases tritium slowly.

The tritium removal system can either be dedicated to a single secondary containment or it can serve several secondaries. A dedicated system can be designed with oxygen and/or water removal to provide an inert glove box atmosphere when desired. The advantage of a common system is that flexibility, redundancy and spare capacity can be built in more easily (Fig. 32).

In the event of a significant release of tritium to the glove box atmosphere, provision for recovery of this tritium and decontamination of the glove box atmosphere is strongly recommended. This is usually done either in a central system serving a number of glove boxes or in a cleanup system dedicated to an individual box. In a central system the flow of gas through an individual box is regulated in such a way as to control the tritium concentration to an acceptable level. In a single box system the gas is normally circulated at a fixed rate. An excellent way to maintain the atmosphere inside the secondary containment or the glove box system is with an inert gas ( $N_2$ , Ar, He, etc.) purge system. A small flow of gas of a few litres per minute can be swept through the secondary containment, which is maintained at about  $-1.5$  cm of water pressure. The continuous purge of gas removes tritium that may

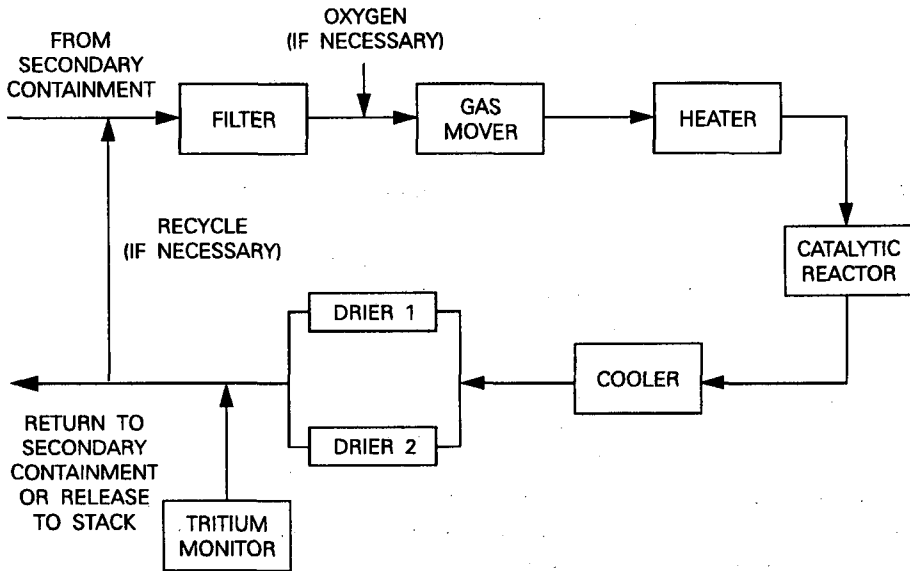


FIG. 32. Oxidation based system to decontaminate secondary containment atmosphere.

leak into the box and provides a sample for a tritium monitor to detect any leakage. If a high concentration of tritium is detected, a higher purge rate can be activated. The discharge gas can be sent to a tritium capture system. In addition, the purge system controls are designed so that glove boxes cannot be overpressurized or underpressurized.

It should be noted that nitrogen is not truly inert and can react with tritium to form ammonia. This ammonia formation is most likely to occur as a result of nitrogen leaking into the process from the secondary containment system.

The most widely used atmospheric cleanup technique is oxidation of tritiated hydrogen to water, followed by adsorption on a molecular sieve bed. This conversion of tritium from a less toxic (HT) to a more toxic chemical form (HTO) means that the removal factor must be very high. In an inert atmosphere system it is necessary either to add oxygen to the oxidation catalyst or to use an oxide containing material such as copper oxide. If oxygen concentration control is important, care must be taken not to allow excess oxygen to pass through the cleanup system back into the box. A schematic flow diagram for an oxidation based system is shown in Fig. 32.

Metal getter based cleanup systems can also be used. Metal getters can only be used with inert gases. They operate either by reaction with the hydrogen to form a hydride or by dissolution of the hydrogen in the metal. Metal getter based systems

also remove other chemical impurities in the gas, such as O<sub>2</sub>, H<sub>2</sub>O, CO<sub>x</sub> and N<sub>2</sub> by reaction with the hot metal. However, the kinetics of tritium absorption by some getters at low concentrations (less than 50 MBq/m<sup>3</sup>) may be inadequate for efficient removal. For these getters to be useful for tritium removal it may be necessary to add hydrogen to the atmosphere to carry and isotopically overwhelm the tritium. Metal getter based glove box atmosphere cleanup systems are available commercially.

The choice of the detritiation system(s) will depend on the radiological hazard potential involved (the amount and chemical form of the tritium) and on technical and economic considerations.

For inert atmospheres the combination of a getter based system in parallel to the standard catalytic oxidation detritiation system is a possibility for those cases where large amounts of tritium gas could be released accidentally from the primary containment. Here one could recover the bulk of the tritium on a getter before switching back to the oxidation/drier operation mode for final cleaning. This technique would minimize the generation of the more hazardous HTO.

Various operational modes for detritiation systems have been satisfactorily employed. One of these is a batchwise treatment of the static box atmosphere each time preset levels of contamination are reached, with subsequent discharge or recycle of the treated gas volume. Another mode is continuous treatment with recirculation of the gas to the box. For normal operation the number of volumes recycled per hour can vary from 0.1 to 2, depending on the leaktightness of the glove box and primary containment systems. When a significant release of tritium occurs in the glove box, the flow may be increased by up to a factor of 20. However, a flow exceeding 20 volume changes per hour is unnecessary in most situations. A disadvantage of multiple glove boxes connected to a central recirculation system is that releases in one box might lead to the contamination of other boxes not primarily involved in the accident.

The detritiation rate increases with the recirculation rate, as do the capital and operating costs. However, high recirculation capacities minimize contamination of the box, especially if part of the tritium is in oxide form, and reduce the risk of release from the glove box to the room.

#### *7.3.3.4. Instrumentation on glove box systems*

The composition of the glove box atmosphere is normally monitored by on-line instrumentation at least for tritium. For inert atmospheres the concentrations of moisture and oxygen may also be measured.

Other necessary instrumentation is a differential pressure control system to prevent abnormal pressure in the glove box, which could destroy the integrity of the glove box. Figure 33 illustrates a typical purge control system for a nitrogen atmosphere glove box that has been used successfully. The system includes flow

meters for control of the nitrogen flow rate, regulators for pressure control and differential pressure switches for measuring pressure inside the glove boxes. The pressure switches shut off either the inlet or the exhaust, depending on which out-of-range condition exists, if the pressure does not fall within preset ranges.

The glove box should also be provided with a passive pressure relief system that operates safely irrespective of whether the box pressure is positive or negative with respect to the laboratory. A system used at several laboratories consists of two passive oil bubblers in parallel. As illustrated in Fig. 34, this simple system operates effectively without allowing oil to be blown out of or sucked back into the box. Vacuum pump oil has proven to be an acceptable oil for these bubbler units.

A typical system has 5 cm copper lines from the glove box through the bubbler to the stack exhaust duct. The total pressure drop from the box to the stack varies with the length of the line, level of vacuum pump oil in the bubbler and stack exhaust pressure. This pressure drop should be approximately 13 cm of water with 8 cm of oil in the bubbler and a 1.7 m<sup>3</sup>/min flow through a 5 cm diameter line. The oil level in the bubbler can be adjusted to reduce the pressure in the glove box to the desired level. Valves in the bubbler line should be avoided.

Flow limiting devices are recommended for any gas or vacuum source entering the glove box. Any sum of these sources, gases or vacuum, should not exceed the flow capability, positive or negative, of the passive pressure relief system for a particular glove box.

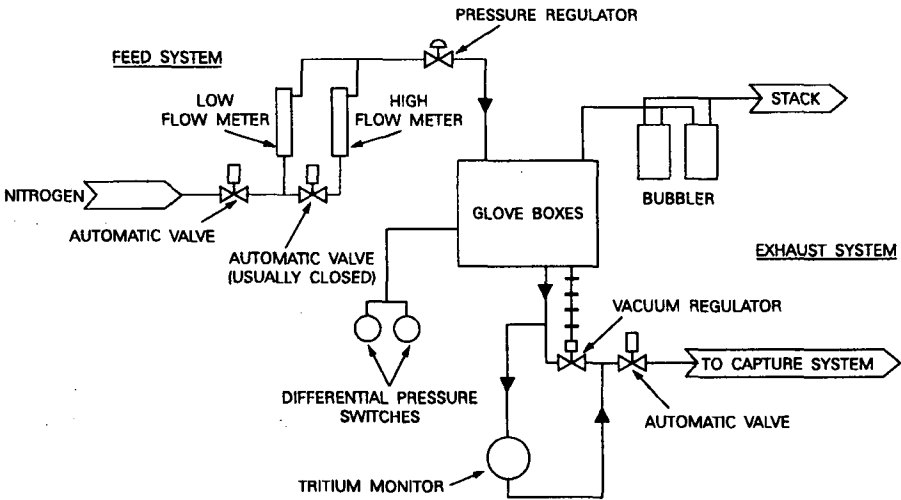


FIG. 33. Purge control system for secondary containment.

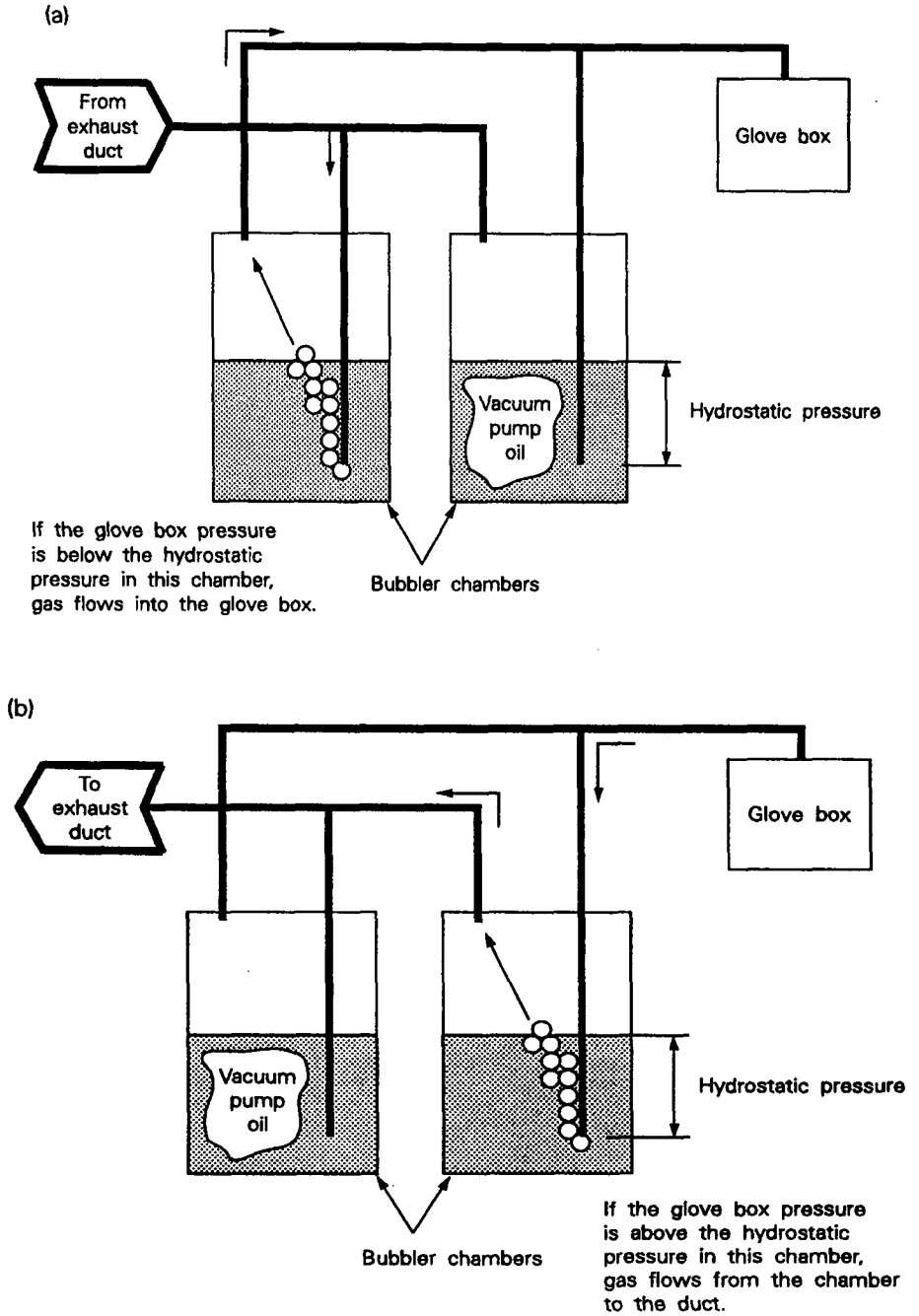


FIG. 34. Glove box line passive relief system. Bubbler operation with glove box pressure (a) low and (b) high.

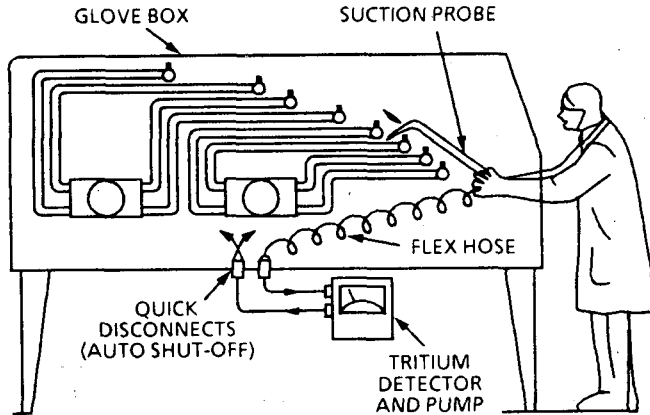


FIG. 35. Leak detection in a glove box with an external pump and tritium monitor.

A simple but very effective and sensitive method of detecting tritium system leaks inside a glove box is illustrated in Fig. 35 [139]. The suction probe, connected to an external flow-through tritium monitor, can pin-point system leaks with high accuracy and sensitivity.

#### 7.3.3.5. Loss of secondary containment

Accidents causing the release of tritium from the process equipment might lead at the same time to the rupture of the secondary containment as a result of a common failure mode. Design and operational provisions should be made to reduce the probability of such events to negligible levels. However, such accidents can happen and would cause the dispersion of tritium into the working area.

A forced ventilation system should be provided in every tritium laboratory to remove tritium from the room, with the exhaust air being monitored before processing or release through a stack to the environment. In order to limit the spread of contamination, the division of the facility into a series of compartments and pressure zones is advisable. These should be arranged so that the more highly contaminated areas are at the lowest pressures. If the secondary containment is breached, two concepts are applicable, i.e. the dilution-release and the confinement-recovery concepts.

In the dilution-release case provisions are made in the plant design to allow an increased flow rate of the room ventilation air in order to purge the contaminated area quickly. This will minimize the exposure of operators and the possibility of tritium sorption by the building and equipment structures.

In the confinement–recovery case the contaminated area is completely isolated and the air is recirculated through a detritiation system similar to that for glove boxes but of larger size. A small fraction of the air exiting the cleanup system is not returned to the laboratory but is released to the exhaust stack. This will allow the contaminated laboratory to be maintained at a pressure below that of the surrounding area. As the size of the cleanup system increases so does its cost, but if it is too small the time required for cleanup becomes unduly long. Some of the released tritium will interact with the room structures and will be sorbed by them. This tritium will cause increased room air contamination by a slow, continuous desorption process. Since any capture process has both risks and benefits, an analysis should be performed of the cost of such a system, the probability of a significant release, the amount of tritium potentially released and whether the benefits associated with protection of the public justify the associated higher exposure levels to operators.

#### 7.4. MAINTENANCE AND CONTAMINATION CONTROL

##### 7.4.1. Introduction

Maintenance requirements and provisions should be major considerations in the design of a tritium facility. The layout of the facility should provide for segregated radiation controlled and uncontrolled zones with ventilation air generally flowing from clean to contaminated zones. Figure 36 shows typical pressure differentials maintained among the different zones of a tritium facility.

		OPERATING LAB. -0.25		
		GLOVE BOXES -1.3		
UNCONTROLLED AREA 0.00	ACCESS CORRIDOR -0.13	MAINTENANCE CORRIDOR -0.4	ACCESS CORRIDOR -0.13	UNCONTROLLED AREA 0.00
		GLOVE BOXES -1.3		
		OPERATING LAB. -0.25		

FIG. 36. Pressure differentials in a tritium facility. All pressures are relative to atmospheric pressure and are expressed in cm of water column pressure.



The interior finish of the walls and ceilings of the radiation controlled areas should be smooth, easy to clean and free of pinholes. Concrete block with filters and epoxy paint has worked well for several facilities. Vinyl floor tile has been acceptable in some facilities, and continuous vinyl has been used in areas where contaminated liquids (water or oils) may be handled and cracks cannot be filled by waxing.

Penetrations for electrical, pneumatic, water and other services into the radiation controlled area, as well as doors and access ways to the area, should be provided with leaktight seals. Design features of these seals have been described [27].

#### **7.4.2. Maintenance of containment systems**

Provision should be made during the design stage to allow maintenance to be performed within the glove box without opening it, to the extent feasible. For example, tritium handling systems can be built so that they can be disassembled within the glove box, and the resulting pieces should then be small enough to fit through the transfer chamber. Systems and components that are vulnerable to failure, e.g. heaters, should be easily accessible so they can be changed without opening the glove box. The maintenance reach must be considered. For example, it may be necessary to reach an item with two hands to get wrenches on fittings and equipment. A good way to guarantee maintainability is to install the maintenance fittings through the glove ports.

If it is necessary to open the glove box to carry out maintenance, the operation should be carefully planned and prepared, particularly if high level tritium contamination is known to exist or is anticipated. Planning and preparation consist of assessment of the radiological hazards, with particular consideration given to the potential presence of tritiated water; reduction of hazard levels if possible; a pre-job conference; a formal radiological work plan or work permit; and provision of personal protective clothing and equipment if they are considered necessary.

An important consideration is local ventilation and contamination control during the 'breaching' operation. At some laboratories it is the practice to extend the envelope of the secondary containment by providing a temporary ventilated 'tent' enclosure over the breached secondary containment. This tent can be set up in a maintenance access corridor common to the back of two rows of glove boxes (Figs 36, 37). With this tent in place, sections of the secondary system can be removed, allowing access to the primary systems. Before the primary or secondary tritium containment systems are breached, all tritium should be pumped to another system and isolated from the area to be breached. After removal of the tritium the system to be breached should be placed under vacuum or purged to a capture system to remove the residual tritium. At the Tritium Systems Testing Assembly (TSTA) at Los Alamos National Laboratory, New Mexico, the use of large diameter, flexible, local ventilation hoses ('elephant trunks') has been found very effective.

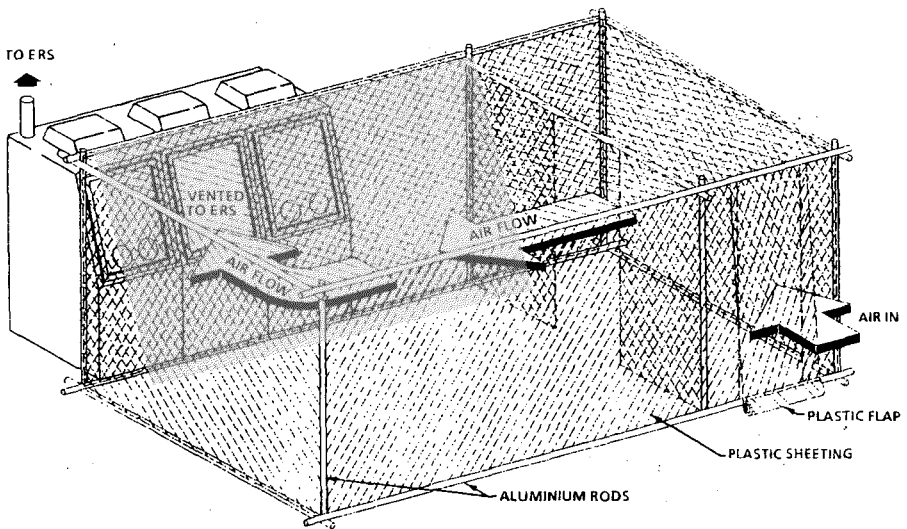


FIG. 37. Use of tent to provide ventilation and contamination control during maintenance operations (ERS: effluent recovery system).

Exhaust from the tent or the elephant trunk can be sent to the stack or, if necessary, to a common air cleanup system for the facility.

Techniques have been developed for transferring major equipment, too large to fit through the transfer chamber, without opening the glove box. One of these designs is described in Ref. [139].

#### 7.4.3. Contamination control and waste handling

Because of tritium's high degree of mobility, tritium contamination can spread quickly to all media and surfaces in a tritium laboratory. Contaminated materials will out-gas HT and HTO, which can spread in air and adsorb on surfaces or be inhaled. One facility, for example, uses the following containment methods for materials out-gassing low levels of tritium:

< 50 MBq/h	Double plastic bags or Saran wrap
50–1000 MBq/h	Paint cans or metal containers
> 1000 MBq/h	Special handling, including use of secondary container and immobilization.

Care must be taken if such out-gassing materials are to be used again at some later date, as the tritium buildup in a closed container may not be known to the unwary worker who has to open it.

(a) Pump oil

In many tritium facilities conventional oil sealed vacuum pumps are still used and the pump oil becomes irreversibly contaminated with tritium. Spent pump oil may be aspirated into a pre-evacuated waste container filled with a sorbent material such as vermiculite or diatomaceous earth. A method of doing this in a safe, 'hands-off' manner has been described in Ref. [140].

(b) Spills

In the event of a spill of water or oil which could be contaminated with tritium, it is important to verify that no skin or clothing is contaminated. Rapid percutaneous absorption takes place if the skin is wetted with HTO, as discussed in Section 2. Known or suspected wetted skin should be immediately washed for 5-10 minutes with soap and water. All contaminated clothing should be promptly removed and bagged. The individual should take a shower as soon as possible.

Spills on floors should be absorbed with clay, vermiculite, silica gel or absorbent paper. Clay is probably the best medium for large spills, particularly for oils and solvents. It is important to ensure that contamination is not tracked to other areas of the laboratory by personnel. Access to the area should be controlled and disposable overshoes should be used if access to the area is necessary. The contaminated area should be mopped or washed with soap and water.

#### 7.4.4. Decontamination

In larger tritium facilities, dedicated decontamination facilities should be provided in the radiation controlled area. These may consist of decontamination glove boxes or fume hoods, with the latter provided with water and/or steam for cleaning and a water collection system.

A simple, effective method of decontaminating the inside of the glove box transfer chamber has been described [139]. If contamination is in the form of HTO the transfer chamber is evacuated and a little water aspirated into it during air back-fill. The surfaces are left exposed to the moisture laden air for several hours to allow isotopic exchange to take place and the transfer chamber is then evacuated again. This process can be repeated until the contamination level is acceptable. For oil contamination a small amount of volatile organic solvent can be aspirated into the chamber instead of water.

If necessary, contaminated metallic equipment may be decontaminated by heating to 300°C for several hours [141].

#### 7.4.5. Tritium storage

There are two methods commonly used for storing HT. These are bulk storage as a gas in tanks and solid storage as a tritide. Bulk storage tanks are the most com-

mon storage units and are the easiest to design. Working pressures near atmospheric pressure are the most common, although some higher pressure applications exist. Any secondary containment system should be designed to withstand a total release of the contents of the primary containment system.

HT is often stored in metal getter beds. This publication is not concerned with the technology of gettering; however, there are certain safety aspects that should be recognized.

Storage of HT as a hydride is common. Several hydrides are available for this use, with uranium hydride being the most common. In a 2 L volume at room temperature with an overpressure (dissociation pressure) of less than 1 torr ( $1.33 \times 10^2$  Pa), 1000 L (STP) of hydrogen can be stored as uranium hydride powder. The hydrogen can be released easily by heating the uranium hydride to 400°C, producing a 1 atm ( $1.013 \times 10^5$  Pa) dissociation pressure. Higher temperatures are required if higher pressures are needed. Temperatures above 700°C should be avoided because uranium can interact with the vessel walls and cause failure of the primary container. Many other metals and alloys form hydrides with different storage characteristics. Hydrogen/metal ratios and dissociation pressure-temperature relationships are of special design interest. Among the metals and alloys used for HT storage are palladium, titanium, lanthanum-nickel, zirconium-nickel and zirconium-cobalt.

Uranium that has been activated by cycles of hydriding and dehydriding is pyrophoric when exposed to air or other oxidizers. Uranium hydride is also mildly pyrophoric. Release of HT from a metal tritide bed is likely to occur if the bed is exposed to air. This potential accident scenario has been studied in Ref. [142]. According to this study the most probable scenario is the opening of a small crack in the tubing, or its joints or fittings, connecting to the storage bed. If this takes place in an air atmosphere glove box, the slow leaking of air into the storage bed would result in oxidation of the surface of the partially hydrided metal.

Other, less likely scenarios could involve the inadvertent opening of an evacuated uranium bed filling line to air or other oxygen containing gas while loading the uranium bed with HT, handling and inspection of returned uranium beds, etc. Breakage of uranium bed filling lines, valves or rupture discs due to a major mechanical impact during handling or transportation is also possible.

Accidental overheating of uranium beds can result in U-Fe reactions, failure of containment and release of tritium as HTO.

Fine particles of uranium tritide deposited within the uranium beds could be exposed to air after opening of the system for maintenance. Experiments [143] have shown that uranium particulates can migrate through the filter of a uranium bed into the system line if filters with porosity exceeding  $5 \mu\text{m}$  are used. Sintered metal filters with a porosity below  $5 \mu\text{m}$  appear to be satisfactory for preventing the carry-over of submicrometre particles of uranium tritide into operational systems.

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## Annex I

### GASEOUS TRITIUM LIGHT SOURCE MANUFACTURING

#### I-1. INTRODUCTION

There is an increasing market for light sources that can operate without a power supply in applications where great reliability is required even in the event of a power interruption. Some current applications are exit signs and aviation landing aids.

Activation of a zinc sulphide phosphor with  $\beta$  radiation will provide such a self-sustaining light source. Tritium in the form of HT gas is an ideal radionuclide for this purpose because, owing to its relatively low radiotoxicity, it can be used in a high specific activity form with reasonable safety, resulting in a high light output per unit volume.

Gaseous tritium light sources (GTLs) usually consist of borosilicate glass capsules or tubes in a variety of shapes and sizes, internally coated with a zinc sulphide phosphor. After coating the capsule is evacuated, backfilled with HT and sealed. The phosphor then becomes activated by the  $\beta$  radiation from the tritium and emits light. Depending on the type of phosphor used, the emitted light varies from ultraviolet to low infrared. The internal pressure of the HT within the capsule can range from about 10 kPa up to about 250 kPa, depending on the brightness requirements.

The typical tritium content of unassembled GTLs varies from 0.05 to about 2500 GBq (0.001 to 60 Ci). Examples of current products in which one or more GTLs are shock mounted together within the assembly are listed below, along with the approximate ranges of tritium content of the devices.

- (a) Aircraft exit signs and markers, 50–500 GBq (1–10 Ci);
- (b) Commercial exit signs and markers, 250–1000 GBq (6–25 Ci);
- (c) Instrument illumination and military products, 50–1000 GBq (1–25 Ci);
- (d) Aviation landing aids, >2000 GBq (50 Ci) in each GTL; assemblies could contain up to 50 TBq (1000 Ci).

#### I-2. DESCRIPTION OF THE MANUFACTURING PROCESS

Commercial HT gas is usually supplied in one of two forms: as HT in a container or as a uranium tritide within a stainless steel container (up to 2000 TBq per container). The second form, uranium tritide, requires that a method of heating be provided to drive the HT from the container.

Most of the current manufacturers of GTLSs transfer the HT from the containers onto uranium beds. The uranium beds then supply HT to the GTLS filling systems.

The GTLS filling systems are usually contained within a ventilated enclosure and are constructed of stainless steel tubing using stainless steel bellows valves to contain the tritium. To reduce inventory, 6.35 mm (0.25 in) o.d. tubing is sometimes used for the tritium gas handling system, while larger tubing is used for the vacuum lines. The vacuum system that is commonly used is a combination of oil seal rotary pumps backing an oil diffusion pump.

The GTLS is usually constructed in such a way that it can be easily connected to the system for evacuation and backfilling with tritium gas. During the evacuation process, heat is commonly applied to the source to remove as much water vapour and other contaminants as possible. When the tritium has been backfilled to the correct pressure, the GTLS is sealed using the heat from a gas/oxygen flame or a CO<sub>2</sub> laser [I-1]. The tritium gas left in the connecting stub and pipework is recovered back to a uranium bed before evacuation and repetition of the process. Laser sealing is more uniform and can be performed in an enclosure under inert atmosphere, but in practice it is limited to maximum tube sizes of about 6 mm o.d. If it is required that the GTLS have tritium pressures above atmospheric pressure, it can be immersed in liquid nitrogen before sealing by gas/oxygen flame. Once removed from the liquid nitrogen, the GTLS returns to room temperature. Pressures of up to 250 kPa (>2 atm) can be obtained by this technique. The sealed GTLS is then decontaminated if necessary and assembled into the finished product.

### I-3. OUTLINE OF SAFETY PROBLEMS

During production of GTLSs it is practically impossible to avoid conversion of some HT to HTO. Since the two chemical forms have very different radiotoxicities (DACs for HT and HTO differ by a factor of 25 000 [I-2]), careful consideration should be given to reducing conversion as much as possible. Where this cannot be done, measures should be developed to reduce exposure to individuals.

The following is a partial list of the specific problem areas.

- (a) Uranium traps,
- (b) Pipework and valves,
- (c) Vacuum pumping systems,
- (d) Surfaces exposed to HT and HTO (i.e. ductwork and enclosures),
- (e) Leakage during seal-off procedures,
- (f) Repair and maintenance.

## I-4. METHODS FOR CONTROLLING EXPOSURE TO TRITIUM

### I-4.1. Double containment

A uranium bed can be used for the storage of HT and its supply to a GTLS filling system. The normal temperature range for the use of such a system is 20–500°C. Care must be taken to prevent the temperature from exceeding that at which the uranium and iron combine, which could result in failure of the container. Even with a thick wall stainless steel container, some of the contained HT will eventually permeate through the wall and be released from the surface in the form of HTO, particularly during the heating cycle. This permeation, in a ventilated enclosure, not only contributes to stack releases but results in surface contamination of the enclosure, equipment and ductwork.

This HTO release can be reduced by the placement of the uranium bed within a secondary containment vessel, usually constructed of stainless steel. Any releases into the containment vessel during normal use of the unit can be recovered by cycling the gas mixture over a uranium bed. A secondary benefit with this type of containment is that a large tritium source would have double protection in the event of an accident.

### I-4.2. Control of tritium leakage during seal-off of the capsule

Two main methods are currently in use for the sealing of the glass tube after backfilling with tritium gas: (1) a manual method using a flame to form a seal; (2) use of a CO<sub>2</sub> laser beam to form a seal.

Leaking tritium gas can be oxidized during the sealing process and methods must be adopted to minimize the exposure to such leaks.

If the flame method is used, inert atmosphere glove boxes are impractical. Therefore, this operation has to be conducted in an atmospheric glove box or a ventilated enclosure. With a ventilated enclosure, consideration might be given to a localized air sweep in the seal-off area to remove the gas mix to a tritium cleanup system or storage container for later cleanup. This system would be activated only when a leak occurs. With an atmospheric glove box, consideration might be given to diverting the ventilation through a tritium gas cleanup system when the leakage occurs. In both cases, a method of closing off the gas/oxygen flow to the seal-off torch is essential to prevent further oxidation of the tritium gas during the leakage.

### I-4.3. Tritium cleanup systems

Tritium cleanup systems are usually based on oxidizing tritium over a hot catalytic bed followed by absorbing the resulting oxide on desiccant driers.

As discussed earlier, the radiotoxicities of HTO and HT differ by a factor of about 25 000; therefore, great care must be taken with tritium cleanup systems that convert HT to HTO.

Use of desiccant driers alone will remove the much more hazardous HTO from a mixture of HT and HTO.

The decision of whether or not to oxidize HT in gaseous effluents to HTO and retain the HTO, which is much more hazardous than the HT, should result from a risk-benefit analysis after considering all the options. The decision will also depend on individual site characteristics, licence requirements and equipment design, and will not be discussed further here.

The most commonly used oxidizing beds consist of a precious metal catalyst on alumina or copper oxide. Operating the bed at a temperature of 500°C will oxidize any tritiated methane generated in the GTLS systems. The drying system usually consists of two molecular sieve beds in series. The addition of water vapour between the two beds will increase the tritium removal from the second stage molecular sieve bed but will also generate a significant amount of low specific activity HTO to be disposed of.

To clean up the exhausts of pumping systems, particularly those with oil (oil seal rotary pumps), it is recommended that an oil recovery-filter system at the exhaust port be used to prevent, as much as possible, the discharge of oil vapours. Useful information on tritium cleanup systems can be found in the literature, for example in Ref. [I-3].

## I-5. MONITORING

It is important to recognize and have the ability to monitor all the potential sources of tritium exposure within the commercial manufacturing environment. These come from the three principal forms of tritium: HT, HTO and tritiated particulates.

Normal plant monitoring techniques including simple, inexpensive systems can be used, for example:

- A HT monitor;
- A HT monitor with a drier between two ionization chambers for a differential monitor;
- A liquid bubbler with a pump and flow meter;
- Dry wipe testing and counting in a  $2\pi$  gas proportional counter or liquid scintillation counter;
- Diffusion or squeeze bottle techniques.

By dividing the tritium handling area into the smallest sections possible and providing sampling points in order that any of the first three systems can be readily connected, procedural monitoring can be accomplished.



## I-6. MATERIALS AND EQUIPMENT SELECTION

Section 5 covers the selection of materials and equipment, but information specific to the GTLS manufacturing process is covered here.

### (a) Valves

The proper selection of valves is important to keep HT under control in a gas handling system and to prevent its leakage to the atmosphere of glove box or containment systems. Stainless steel bellows valves with metal to metal seals, which are either welded or connected to the system with metal gasket connectors, have been used successfully, although metal seals have been known to leak. If metal to metal seals are for some reason not suitable, polyimide or Kel-F stem tips may be used. Caution is recommended in the use of Kel-F, owing to the generation of tritiated hydrogen fluoride and methane. The typical lifetime of this type of seal when exposed to HT at atmospheric pressure for seven hours a day, five days a week, is 12 to 18 months. Therefore, for safety reasons, it is recommended that seals be changed every six months.

### (b) Vacuum pumps

Vacuum pumping systems are now becoming available that operate with small amounts of special oil or entirely without oil. As much as possible, oil should be avoided in GTLS systems.

### (c) Materials

The use of either 316 or 304 stainless steel with either welded or silver brazed joints is recommended. Internal cleanliness is essential, and internal surfaces should be free of inclusions and have a smooth finish.

## I-7. TRITIUM SAFE HANDLING PROCEDURES

The following are procedures used by a particular manufacturer in a tritium safe handling programme.

### I-7.1. Zoning for contamination control

Zoning should be set up to restrict the access to areas where HT is being handled and to prevent the spread of contamination to areas where HT is not handled.

Zone room pressures should be such that the flow of air is directed toward the more contaminated area. Acceptable control limits for airborne and surface tritium contamination are:

- (a) Zone 1 — tritium gas handling area
  - (i) Airborne contamination not greater than  $0.5 \text{ MBq/m}^3$  ( $10 \text{ } \mu\text{Ci/m}^3$ ), monitor alarm set at  $0.2 \text{ MBq/m}^3$  ( $5 \text{ } \mu\text{Ci/m}^3$ );
  - (ii) Removable surface contamination not greater than 3000 dis/s per  $100 \text{ cm}^2$  wipe.
- (b) Zone 2 — sealed source handling area
  - (i) Airborne contamination not greater than  $50 \text{ kBq/m}^3$  ( $1 \text{ } \mu\text{Ci/m}^3$ );
  - (ii) Removable surface contamination not greater than 80 dis/s per  $100 \text{ cm}^2$  wipe.

When the limits of any zone are exceeded, action is taken immediately to correct the problem and prevent exposure of personnel and release to the environment. Urine samples are collected and analysed weekly from all workers in the two zones or following suspected exposures.

### **I-7.2. Personal protective equipment**

All personnel should wear at least the following protective equipment:

- (a) Laboratory coat (Tyvek type, polyethylene coated),
- (b) Rubber overshoes,
- (c) Surgical or PVC gloves.

Change procedures should be implemented when personnel enter or leave the area and a sink should be provided for washing of hands.

Under certain conditions, such as maintenance or repair operations, personnel may be required to wear the following protective equipment:

- (1) Coveralls (Tyvek type polyethylene coated),
- (2) Rubber overshoes,
- (3) Hood (over hair),
- (4) Face mask,
- (5) Two pairs of surgical or PVC gloves (top pair changed frequently during operation).

### **I-7.3. Maintenance and repairs**

When a HT handling system is exposed to air during a repair or maintenance operation, large quantities of HTO can be generated. If the repair or maintenance

operations are conducted within a total containment glove box, this is less of a problem; otherwise significant releases and exposures can occur. Restriction of the exposures and releases can be achieved by using one or more of the following methods.

- (a) Repeated flushing of the HT handling system with hydrogen and absorbing the gas on a uranium bed.
- (b) Repeated flushing of the HT handling system with helium gas through a tritium cleanup system.
- (c) After the use of method (b), atmospheric air is allowed to enter the HT handling system. The system is closed for a period of at least 30 minutes and the air from the HT handling system is then pumped through a tritium cleanup system and method (b) is repeated. This operation can be conducted several times to restrict the release to an acceptable level.
- (d) Placing a plastic bag type glove box around the component to be repaired or replaced and flushing the bag with an inert gas such as argon, through a tritium cleanup system.

#### **I-7.4. Safety committee**

Safety committees have been effective in increasing safety. Such a committee typically consists of senior staff members from the following areas:

- (a) Management,
- (b) Engineering (usually design and maintenance together),
- (c) Radiation health and safety (including the radiation safety officer),
- (d) Production.

Under normal operating conditions representatives from all groups form a committee to discuss and inform other groups of current situations. It is recommended that the meetings be conducted monthly. Also, before any unusual operation the committee should meet to discuss all aspects of the operation, such as:

- (1) Type of operation,
- (2) Expected tritium exposure,
- (3) Possible unexpected tritium exposure,
- (4) Protective clothing,
- (5) Assistance, if necessary, from other groups,
- (6) Expected doses,
- (7) Waste disposal,
- (8) Duration of operation,
- (9) Containment,
- (10) Selection of director of operation (for planning and reporting),
- (11) Previous experience.

Other responsibilities of the safety committee would be:

- (i) To review urinalysis results,
- (ii) To review procedural developments,
- (iii) To review licensing requirements,
- (iv) To review the inventory of tritium materials,
- (v) To conduct inquiries into unusual situations or accidents.

## **REFERENCES TO ANNEX I**

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- [I-2] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, Limits for Intakes of Radionuclides by Workers, Pergamon Press, Oxford and New York (1979).
- [I-3] RHINEHAMMER, T.B., LAMBERGER, P.H. (Eds), Tritium Control Technology, Rep. WASH-1269, US Govt Printing Office, Washington, DC (1973).

## Annex II

### COMMERCIAL RADIOCHEMICAL MANUFACTURING

#### II-1. INTRODUCTION

The main commercial uses of tritium, other than in the manufacture of gaseous light sources, are applications of tritiated self-luminous compounds, and the labelling of chemicals and pharmaceuticals. In this annex the tritium handling practices and experience of one manufacturer are described.

#### II-2. MANUFACTURE OF TRITIATED SELF-LUMINOUS COMPOUNDS

The first commercial application of radioactive material was in the use of radioactive self-luminous compounds. With the availability of tritium, with its relatively low radiotoxicity, efforts have been made to develop high specific activity tritiated materials as an energy source for coating zinc sulphide crystals.

This process of coating zinc sulphide is known as luminizing. Tritiated polymers are often used for this purpose with specific activities up to 20 TBq/g (~ 500 Ci/g). Radiolysis, depolymerization and the instability of the C-H bond lead to degradation, oxidation and hydrogen isotope exchange with ambient air and moisture to produce HTO and volatile tritiated organic compounds. Efforts to stabilize these materials have reduced releases of tritium from the compound to the ambient air to 5-8% of the total activity per year. About 90% of the out-gassed tritium will be in the form of HTO; therefore good radiation protection efforts must be made.

#### II-3. EXPOSURE CONTROL IN THE LUMINIZING INDUSTRY

In the control of occupational tritium exposure in luminizing operations, three main exposure routes should be considered:

- (a) Tritium luminous powder must be handled as an open radioactive source. The fine grain powder (10-40  $\mu\text{m}$ ) is easily spread or blown around.
- (b) Powder material, in contact with air and humidity, releases some 5-8% of its activity per year.
- (c) To apply tritiated luminous compounds (e.g. to dials), they must be mixed with some transparent binder containing volatile solvents. Tritium exchanges with these solvents, which subsequently evaporate into the air.

Tritium luminous compounds now have been used for more than twenty years. Experience has shown that the following practices are useful in controlling occupational exposure.

- (1) Luminizing rooms should be clearly marked with radioactivity symbols and designated as controlled areas.
- (2) Luminizing rooms must be used only for applying luminous compounds.
- (3) Floors should be covered with welded plastic flooring extending 10 cm up the walls for easy decontamination. Walls should be washable and working areas covered by smooth, easy to clean surfaces.
- (4) Furniture should be reduced to an absolute minimum.
- (5) Luminizing rooms should be ventilated with an air exchange rate of at least five times an hour.
- (6) Room air exhaust should be filtered to remove particulates before being sent to the stack.
- (7) Work bench ventilation must withdraw all HTO vapour as completely as possible.
- (8) Drying should be performed in a ventilated oven or box.
- (9) Luminous tritium compounds should be stored in filtered glove boxes that are kept at a minimum negative pressure of 5 mm water column. Separate exhaust piping is required in order to ensure isolation from room ventilation.
- (10) The inventory of luminous compounds should be minimized.
- (11) Transfer of contamination from controlled to non-controlled areas can be reduced by personnel washing their hands and changing their shoes and overcoats. Inspection under UV light will reveal the presence of luminous compounds.
- (12) Floors of luminizing rooms should be washed at least once a week.
- (13) Eating, drinking and smoking must be forbidden in controlled areas.
- (14) Luminous compounds may be mixed only in a glove box.
- (15) Instructions for the proper handling of tritium luminous compounds must be drawn up and workers trained.
- (16) Luminizing operations are to be declared as 'Working Condition A', as defined in the IAEA Basic Safety Standards [II-1]. Individual assessment of effective dose equivalent shall be carried out for all workers by means of urinalysis.

By following these practical recommendations and directions, it has been possible to reduce exposures in the luminizing industry substantially even though a worker may handle 150 g of tritium in a year. Figure II-1 shows the average annual effective dose over the period 1969-1986 for one company.

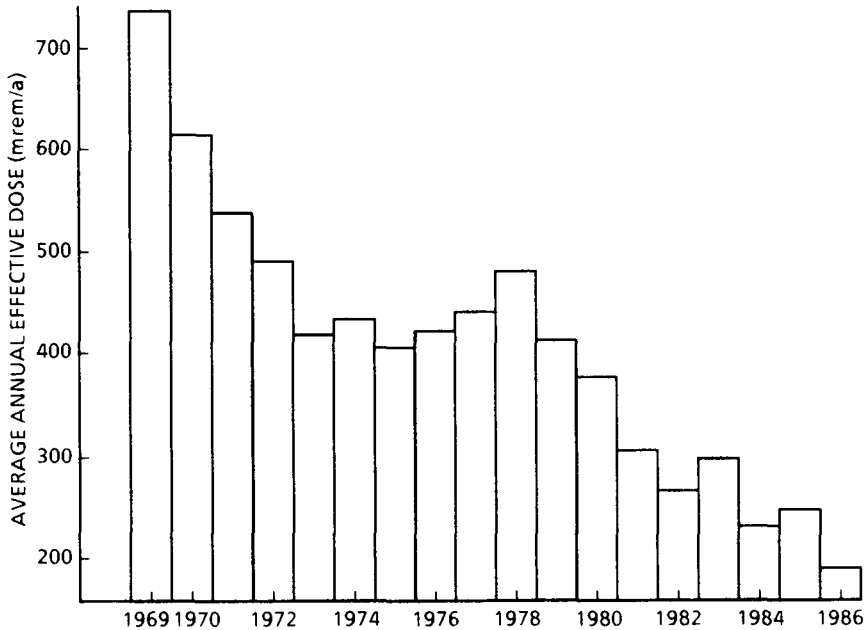


FIG. II-1. Average annual effective dose to workers in a luminizing company.

#### II-4. TRITIUM LABELLING

Tritium labelling operations where more than 500 GBq (10 Ci) are used should be performed in controlled laboratories with forced and monitored air exchange of about 10 times per hour. The rooms should be kept at a pressure of at least 5 mm of water column lower than the surrounding uncontrolled areas. The tritium operations themselves should be performed in ventilated glove boxes with an air exchange rate of 20 times per hour and with the pressure kept at 50 mm water column lower than the room. All HT can be handled in these boxes in a primary containment system, which is a stainless steel system consisting of tubing, metal bellows valves with stainless steel seats and stems, and metal to metal seals. Such a system has the great advantage that most organic contamination, water and solvents can be removed by heating the whole system to high temperatures. In some systems, the reaction vessel is made of Pyrex glass, which is fitted with tritium compatible connections to the all-metal main system.

Fresh HT can be generated from uranium beds having 3 mm thick 316 stainless steel walls to minimize diffusion during the heating process. Tritiation pressures can rise to 170 kPa. Unused HT can be reabsorbed by recirculation over a flow-through uranium trap by means of a toepler pump to increase absorption speed, because  $^3\text{He}$

is less likely to block the uranium surface than in one way traps. Residual gas can be evacuated with turbomolecular pumps to achieve a good vacuum and prevent back-diffusion of oil vapour from two-stage oil rotary forepumps into the systems. The exhausted tritium containing gases are passed from the pumps over a molecular sieve and activated charcoal filter into storage tanks. After the tank pressure reaches 95% of the ambient atmospheric pressure, the contents are recirculated through a catalytic converter and molecular sieve columns until the tritium concentration is reduced to an acceptable level. The gas can then be released to the atmosphere.

With a containment system such as that described above, tritium concentrations in urine of workers in the labelling area should be maintained below 100 kBq/L, which results in an annual committed effective dose equivalent of less than 1.5 mSv.

## II-5. WASTE MANAGEMENT

Organic liquid waste may be adsorbed on kieselguhr enclosed in a glass or plastic container and fixed in a concrete matrix in a welded steel cylinder which is tested for leaktightness by immersion tests. Solid waste of several tens of gigabecquerels per kilogram is compressed and also welded in steel cylinders. Intermediate wastes should be stored in glove boxes or fume hoods, depending on the out-gassing rate and levels of activity. Highly out-gassing material may be sealed in plastic, embedded in polyurethane or epoxy resins and canned.

## II-6. CONTROL OF RELEASES TO THE ENVIRONMENT

The glove box and room exhaust air should be continuously monitored. Experience shows that only HTO has to be taken into consideration, since a maximum of only 20% of the activity occurs in the form of HT. Authorized release limits are set by the regulatory authority. Confirmation of acceptable releases may be obtained by an appropriate programme of environmental monitoring.

### REFERENCE TO ANNEX II

- [II-1] INTERNATIONAL ATOMIC ENERGY AGENCY, Basic Safety Standards for Radiation Protection, 1982 Edition, Safety Series No. 9, IAEA, Vienna (1982) 172 pp.



## **Annex III**

### **SAFE HANDLING OF TRITIUM IN LOW AND MEDIUM LEVEL UNIVERSITY AND MEDICAL FACILITIES**

#### **III-1. INTRODUCTION**

The IAEA Recommendations for the Safe Use and Regulation of Radiation Sources in Industry, Medicine, Research and Teaching [III-1] give general guidance on the establishment and operation of an appropriate radiation protection programme for use in industrial and medical facilities. An effective and optimized radiation protection programme is illustrated in Fig. 1 of that publication.

#### **III-2. ORGANIZATION AND MANAGEMENT**

It is essential in any operation involving radionuclides that appropriate provisions be made for radiation protection. This involves a commitment on the part of senior management to providing the necessary resources so that all work, experiments and medical procedures are carried out in such a way that all exposures are kept as low as reasonably achievable.

Management shall ensure that all staff are aware of this commitment, that all applicable regulations and procedures are complied with and that all authorities and responsibilities are clearly documented and publicized.

Most large hospitals and universities have a radiation protection officer who assists management in carrying out their responsibilities for radiation protection.

#### **III-3. STAFF SELECTION AND TRAINING**

Appropriate criteria should be established for the selection and recruitment of staff in terms of knowledge and attributes that will have a bearing on their safe working habits.

Training programmes must be established for all workers who will handle tritium in order that they will carry out their duties in a manner such that radiation exposures to patients, members of the public, other workers and themselves are kept as low as reasonably achievable.

The radiation protection officer is a person whose selection and training could have a vital influence on the safe working habits of all staff members and therefore special care must be taken in the selection and training of this person.

## III-4. OCCUPATIONAL RADIATION CONTROL

### III-4.1. Source control

The first principle of radiation control is control of the source. Obviously the hazard of an operation will increase with the activity handled, and careful thought should be given to the question of how much activity is really needed in order to carry out a given experiment or procedure. The temptation to buy more than is needed because the cost per unit activity is lower should be resisted. The quantity of tritium in stock or in use should be the minimum necessary for the procedure or experiment.

When the activity of a batch of stock material is high, it is often possible to subdivide it at an early stage, so that the activity subsequently handled at any one time is reduced.

High specific activity will, in general, increase the hazards, and dilution should be carried out at an early stage if possible.

Stock solutions should be well marked, such that everyone will be aware of their contents. Waste should be removed from the working area as soon as possible and disposed of, as discussed in Section 6.

Work with tritium should be segregated from inactive work in order to reduce the number of persons at risk and to reduce the risk of cross-contamination.

### III-4.2. Design features

Provision of adequate ventilation should be included in the original design of the premises. Routes of entry and exit for the ventilating air should be clearly defined under all conditions of use, including open and closed positions of doors and windows and the various operating arrangements of the fume hoods. In small laboratories it may be possible to provide the needed flow of air simply by the exhaust system of the fume hoods, but in such a case special attention must be given to the inflow of fresh air into the laboratory under all conditions by such means as providing adequate louvres in the doors of rooms. Containment of tritiated liquids by the use of fume hoods is recommended except when only minimal quantities are being handled (a few tens of megabecquerels).

It is important that any spills of tritiated liquid be quickly and thoroughly cleaned up. Bench tops and floors made of smooth, impervious material will greatly facilitate the decontamination process.

### III-4.3. Administrative controls

With respect to the classification of laboratories, the most recent international recommendations are those of ICRP Publication 25 [III-2], which recommends three

TABLE III-1. MAXIMUM LEVELS OF TRITIUM FOR OPERATIONS IN LOW AND MEDIUM LABORATORIES

	Low	Medium
Simple storage	< 1 Ci (<50 GBq)	1-1000 Ci (50 GBq-50 TBq)
Simple wet operations	< 100 mCi (<5 GBq)	100 mCi-100 Ci (5 GBq-5 TBq)
Normal chemical operations	< 10 mCi (<500 MBq)	10 mCi-10 Ci (500 MBq-500 GBq)
Complex chemical operations	< 1 mCi (<50 MBq)	1 mCi-1 Ci (50 MBq-50 GBq)

categories of laboratory — low, medium and high. When the only radionuclide being used is tritium, the ICRP recommends maximum quantities for the low and medium categories as shown in Table III-I.

These values are at least one order of magnitude more restrictive than those recommended in Section 7 of the present publication. Since many university and medical facilities may be using tritium in the form of HTO or organically labelled compounds with a higher radiotoxicity than HT, this greater restriction is justifiable.

Pipetting by mouth or other activities that could increase the risk of ingestion of tritiated liquids must always be prohibited in laboratories where tritium or other radionuclides are handled.

Tritium laboratories should contain only that equipment and furniture necessary for the operations to be carried out. Cluttering work areas with unnecessary equipment or poor housekeeping will increase the risk of accident and make decontamination more difficult.

Protective clothing in low and medium level laboratories need not exceed a simple laboratory coat or overalls and plastic gloves. A plastic apron will give added protection in the event of splashes of HTO.

Before leaving the laboratory, workers must remove protective clothing and wash their hands thoroughly. Protective clothing should be laundered frequently.

### III-5. RADIATION CONTROL FOR THE PUBLIC

Exposure of members of the public should remain trivial as long as the waste disposal practices described in Section 6 are complied with.

### III-6. ACCIDENTS AND DECONTAMINATION

Most accidents in university and medical facilities will involve only small quantities of tritium and therefore will not constitute a major emergency.

Spills should always be cleaned up as soon as possible but, as a general rule, decontamination of persons will take priority over decontamination of working areas. However, steps should be taken at an early stage to minimize the spread of tritium to other areas, e.g. by erecting temporary barriers and warning notices and by restricting the number of persons entering and leaving the area. In the event of a substantial release of HT or a spill of volatile tritiated liquid, the laboratory should be temporarily closed off, with the ventilation left running.

For the decontamination of persons the following procedures should be followed:

- (a) Remove contaminated clothing immediately. HTO uptake will begin immediately and continue as long as contaminated clothes are in contact with the skin. In the event of being severely wetted with HTO, the person should immediately shower (removing clothing in the process).
- (b) Wash hands thoroughly with soap and cold water, scrubbing lightly with a soft brush.
- (c) Wash out any cuts with plenty of water, encouraging gentle bleeding.
- (d) If necessary, wash out eyes with normal (9 g/L) saline, using an eye bath. Distilled or tap water from a standard plastic wash bottle will suffice in an emergency.
- (e) Wash out the mouth with plenty of water.

A urine sample should be collected 2-3 hours after the accident to assess uptake.

Spills should be cleaned up by wet methods as far as possible, to avoid resuspension of dust. Discarded bench coverings, gloves, cleaning materials, etc., should be disposed of as active waste. The use of solvents should be avoided where possible because of their volatility and possible chemical toxicity.

Decontamination of glassware used for active work may be effected by the use of chromic acid or a suitable detergent, but tritium adheres very strongly to glass and it may be difficult to remove the last traces from apparatus used for high level work.

Depending on its chemical form, tritium may be absorbed into exposed materials, and decontamination of the surface is likely to be followed by replacement of tritium by diffusion from below.

All non-trivial accidents involving dispersal of active material should be recorded and investigated and corrective measures taken.

Further guidance on safe handling of tritium in low and medium level laboratories is given in Ref. [III-3].

## **REFERENCES TO ANNEX III**

- [III-1] INTERNATIONAL ATOMIC ENERGY AGENCY, Recommendations for the Safe Use and Regulation of Radiation Sources in Industry, Medicine, Research and Teaching, Safety Series No. 102, IAEA, Vienna (1990) 98 pp.
- [III-2] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, The Handling, Storage, Use and Disposal of Unsealed Radionuclides in Hospitals and Medical Research Establishments, Publication 25, Pergamon Press, Oxford and New York (1977).
- [III-3] MARTIN, E.B.M., Health Physics Aspects of the Use of Tritium, Occupational Hygiene Monograph No. 6, Science Reviews Ltd, Leeds, UK (1982).

## Annex IV

### TRITIUM SAFE HANDLING IN HEAVY WATER REACTORS

Operational experience at Ontario Hydro's CANDU type heavy water reactors is summarized in this annex.

#### IV-1. SOURCE CONTROL

The most effective source control is removal of the source. This is being done for CANDU reactors in Canada by means of dedicated HTO removal facilities, such as the one located at the Darlington Nuclear Generating Station. These facilities extract tritium from tritiated heavy water and convert it to T<sub>2</sub>, which is immobilized and stored in metal tritide beds.

Other HTO source control measures, in approximately the order of their effectiveness, are as follows.

##### IV-1.1. Leaktightness of process systems

The first step in leakage reduction is to eliminate as many joints, or even as many components, as possible, since leakage is a function of the number of joints and components. Systems should be designed to be as simple as possible. However, due consideration should be given to maintenance requirements.

Process systems containing HTO must be designed to much more stringent leak specifications than conventional steam, water or oil systems. Adherence to accepted nuclear design (ASME III Code) requirements, with good quality control, will not ensure a leaktight system. The ASME III Code is concerned with failure; leaktightness can only be as good as the components themselves, which should be specifically selected for HTO service. Manufacturers of mechanical equipment often have little or no experience with the leaktightness standards required for HTO control.

Flanged joints with elastomeric gaskets are particularly prone to leakage and should be replaced with welded joints wherever possible. Joints with metal knife edges and metal gaskets can be used when a flanged joint is necessary. In addition to proper flange design and installation, techniques that could be used to improve leaktightness include [IV-1]:

- (a) Seal welding, which requires the use of specially designed flanges with a claw type seal that can be welded;
- (b) Live loading, which involves loading the studs of a flanged joint to counteract the expansion and contraction of the joint during temperature and pressure excursions.

Mechanical couplings have fundamental leakage problems and should generally be avoided except for low temperature (50°C) and low pressure service. Leakage can be reduced by seal welding.

Valves with stem packings generally leak around the stem. The leakage is reduced somewhat by live loading. Essentially zero leakage valves such as bellows sealed valves, diaphragm valves and magnetically coupled valves are much more satisfactory for handling HTO.

#### **IV-1.2. Removal of HTO from air**

HTO vapour in air is removed by means of desiccant driers. The drier beds are periodically regenerated and HTO is removed as condensate. The most common type of desiccant is zeolite (a molecular sieve), which exhibits greater moisture capacity at low relative humidities than other types of desiccants. Different drier designs, arrangements and regeneration circuits developed for CANDU type reactors have been described [IV-1]. The cost of the cleanup system is related to the size of room that is serviced.

#### **IV-1.3. Confinement of tritiated air**

Localized confinement of systems containing HTO will reduce the spread of HTO contamination, reduce the size of ventilation systems required and make vapour recovery economically justifiable. Small systems can be confined in a small enclosure such as a fume hood or glove box. Larger systems may be confined in small rooms. Design features of confinement rooms as well as leaktight penetration seals for piping and cables have been described in Ref. [IV-1].

#### **IV-1.4. Purge ventilation**

Good ventilation control should prevent the contamination of clean areas by HTO in air from contaminated areas. In large, open buildings such as the reactor hall or turbine hall, significant problems may be encountered with maintaining the general direction of air flow from clean to contaminated areas.

Purge ventilation without air cleanup is a quick way of reducing HTO contamination in a work area, thereby facilitating personnel access. However, this may result in significant releases to the environment, resulting in exposure of the public, and is not generally recommended.

Sometimes local ventilated canopies over components that are known to leak, or ventilated plastic tents over contaminated equipment, can be effective in reducing the HTO hazard to operating personnel and reducing the spread of contamination. Ventilated 'elephant trunks' can also be effective for local containment over contaminated equipment.

TABLE IV-1. EFFECTIVENESS OF HTO CONTROL MEASURES

Control measure	Reduces occupational exposures	Reduces environmental emissions
HTO removal	Yes	Yes
Leaktightness	Yes	Yes
Vapour recovery	Yes	Yes
Confinement and local control ventilation	Yes	No
Purge ventilation	Yes	No

The effectiveness of various HTO control measures is summarized in Table IV-1.

#### IV-2. CONTAMINATION AND EXPOSURE CONTROL

HTO hazards at nuclear facilities result from:

- Chronic HTO leakage from process systems and equipment;
- Acute leakage or spills due to equipment failure and/or human error;
- HTO contamination from maintenance activities.

Practices and procedures for controlling the spread of HTO contamination and occupational HTO exposure are presented below.

##### IV-2.1. Access control

Certain areas in nuclear facilities are subject to hazardous conditions, including high HTO levels, depending on operating conditions or the systems contained in the areas. These areas should be defined and access to them strictly controlled. Access control can be implemented by physical barriers such as access keys, hold-off tags and interlocks, or by administrative barriers such as access permits.

##### IV-2.2. Zone control

The facility should be divided into control zones (typically three) to separate relatively clean areas from contaminated ones. Zoning facilitates the location and



segregation of systems and equipment, and the control of the movement of people and equipment and the spread of contamination. Ventilation generally flows from zones of low to high contamination.

Within the contaminated zone, it is sometimes desirable to set up 'rubber areas' to localize areas of particularly high contamination. These are roped off areas within which colour code rubbers are worn over normal work shoes.

#### **IV-2.3. Procedural control**

Procedural controls such as signs, radiological work plans, the issuing of permits for specific tasks ('safe work permits'), etc., are effective in controlling exposure. The radiological work plan or the safe work permits can, for example, identify clearly the HTO dose targets for a particular job, the estimated HTO contamination levels, the required personal protective equipment and the planned duration of stay in the work location.

#### **IV-2.4. Tritium monitoring**

Lightweight, portable tritium monitors can be used for general surveys, particularly where localized HTO concentrations near process equipment are expected to vary significantly with time during a particular job. Portable monitors can stay with the individual close to the work area, where localized HTO concentrations may be quite different from the average room air concentrations indicated by fixed air monitors. The monitor should include high level alarm functions with at least one audible alarm. Ideally, the monitor should also have adequate  $\gamma$  compensation, except in areas where the  $\gamma$  fields are low or absent.

Fixed area monitors provide a continuous indication of the gross average HTO concentration. They help prevent overexposures by providing an automatic alarm when HTO concentrations exceed acceptable levels. They also permit continuous monitoring of HTO concentrations in various areas without personnel entry and provide prompt information for work planning. System designs using remote, centrally located tritium monitors with sampling lines running from the central location to various work areas have been tried. In principle such systems would reduce the number of monitors required and also permit them to be located in low background areas. In practice these are high cost systems that also have operational problems such as condensation and memory effects in the sample lines.

For many applications where real time readout is not essential, simple bubblers or samplers using solid adsorbents are recommended. They are inexpensive, reliable and relatively free from interference by  $\gamma$  radiation and radioactive gases.

#### **IV-2.5. Dosimetric control**

Urine bioassay measurements not only provide data for the official internal dose record (compliance dosimetry) but are also used in most nuclear facilities for internal dose control, e.g. for provision of prompt dose data to field personnel for the purpose of controlling and limiting HTO doses to planned levels during a given job. To provide effective dose control, the delay between the submission of a urine sample and the return of dose data from the bioassay laboratory should be acceptably short. It would be desirable to have 'direct reading HTO dosimeters' which can be read in the field. Various methods have been tried such as saliva analysis and personal samplers, but no satisfactory method has yet emerged.

Bioassay control limits may be set to restrict work or to remove individuals from radiation work. These limits vary from facility to facility. At Ontario Hydro, for example, the restriction level of HTO in urine is  $25 \mu\text{Ci/L}$  ( $9 \times 10^5 \text{ Bq/L}$ ) and the removal level is  $50 \mu\text{Ci/L}$  ( $1.8 \times 10^6 \text{ Bq/L}$ ).

#### **IV-2.6. Personal protective equipment**

Design and performance aspects of personal protective equipment have been discussed in Section 4. At heavy water based nuclear plants, use of air supplied suits is regular and reusable suits are justified. These could typically be reused about fifty times. The protection factor of suits developed by Ontario Hydro exceeds 1000. For the purpose of dose estimates for work planning, a protection factor of 100 is normally assumed.

Shower facilities should be provided near the exits from work locations to permit showering to remove surface contamination from the suit before undressing.

Air supplied masks or respirators are sometimes useful for quick entries into areas where working time has to be strictly limited because of high  $\gamma$  fields. Since HTO will diffuse through the skin, the effective protection factor when only a respirator is worn is about 2 for HTO.

Gloves must be worn when handling HTO or working with HTO contaminated equipment. Glove materials have been discussed in Section 4.3.

#### **IV-2.7. Dose commitment reduction methods**

Following a significant acute HTO exposure, the committed dose can be reduced somewhat by prompt showering after exposure and by increasing the normal water intake rate. Details of these methods are given in Section 2.4.

### IV-2.8. Control of tritiated water spills

The handling of HTO requires special precautions as the uptake of HTO from wetted skin is very rapid, at a rate in excess of 0.2 g of HTO per square metre of skin per minute [IV-2]. Air supplied suits and gloves should normally be used when handling high concentration HTO. In the event of a spill and the bare skin becoming wetted, the skin should be thoroughly washed with water. All contaminated clothing should be removed and placed in plastic bags. The wetted skin area should be wiped off and a shower should be taken as soon as possible.

Small spills of HTO on the floor can be cleaned up and recovered manually by means of sponge mops. With larger spills, a wet vacuum cleaner can first be used to recover the bulk of the spill quickly before final sponge mopping. Personnel involved in the cleanup should be dressed in air supplied plastic suits. HTO contaminated mops and materials should be placed in sealed double plastic bags for disposal.

Large spills of HTO within containment and confined areas can be recovered in the station's liquid collection and vapour recovery drier systems.

Spills may also be absorbed on media such as absorbent clay and vermiculite. Absorbent clay is probably best for larger spills, particularly tritium contaminated oils and solvents.

### IV-3. TRITIATED WASTE HANDLING

A wide variety of tritiated contaminated solid wastes are generated in heavy water nuclear stations, for example clothing, discarded plastic suits, gloves, mops, vermiculite, floor coverings, filters, ion exchange resins, hardware, piping and tools from maintenance activities. Wastes should be segregated at collection locations into combustible, compactible, and non-processable categories for later efficient processing. Tritiated wastes should be contained in plastic bags to minimize tritium out-gassing into accessible areas. Wastes with high concentrations of HTO should be stored in fume hoods or under ventilated canopies. In general, equipment rooms containing high HTO content heavy water systems should have ventilation air driers.

Spent ion exchange resins used in water cleanup circuits can contain significant amounts of residual HTO. They should be dewatered or detritiated and stored in sealed disposable ion exchange vessels. Spent filter cartridges should be dewatered and placed in sealed metal 'drip cans' to control contamination spread. Packaging and shipment off-site should conform to accepted national waste shipment standards.

Some tritiated wastes should be conditioned and immobilized before storage or disposal. Such wastes could include aqueous wastes, organic wastes and solid wastes. Packaging and conditioning details are given in Section 6.

## REFERENCES TO ANNEX IV

- [IV-1] WONG, K. Y., et al., Canadian Tritium Experience, Canadian Fusion Fuels Technology Project, Canadian Centre for Fusion Fuels Technology, Mississauga, Ontario (1984)
- [IV-2] OSBORNE, R. V., Intake of tritium when skin is splashed with heavy water, *Health Phys.* **15** (1968) 155-156.

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
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