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VARIOUS DESIGN AND OPERATIONAL ASPECTS OF ETHEL

G. Vassallo
European Commission, Safety Technology Institute
JRC-Ispra Site, I-21020 Ispra (VA), Italy
Tel. +39-332-789325, Fax +39-332-785835, E-mail (internet) gary.vassallo@cen.jrc.it

ABSTRACT

The design and operation of a facility handling macro quantities of tritium raise problems not normally encountered in hot laboratories. This paper highlights some anticipated complications with a new tritium facility and discusses how such difficulties can be resolved.

INTRODUCTION

Energy is released when nuclei of light elements fuse together to form heavier ones. The easiest fusion reaction to achieve is that between two isotopes of hydrogen, namely deuterium and tritium[1] [1]:

$$^{2}D + {}^{3}T \longrightarrow {}^{4}He + {}^{1}n \qquad (1)$$

The use of tritium as a basic fuel material in a thermonuclear fusion reactor raises safety issues due to the combined effects of its physico-chemical characteristics and its radioactive nature. In particular, the ability to achieve further significant progress with regard to tritium burning is directly coupled to the demonstration of handling macro-quantities of tritium, say tens of grammes, in a safe and reliable manner. Moreover, containment is a major concern owing to the mobility of tritium and the wide abundance and diffusion of its protium counterpart; effectively, such protium represents an irreversible sink for tritium. Hence, much effort is required to study the behaviour and impact of tritium on technical processes and plant. In addition, research must address tritium in all its forms (gaseous, liquid and solid) throughout the entire concentration range, if possible tritium loss mechanisms and pathways are to be identified and tackled.

The European Tritium Handling Experimental Laboratory (ETHEL) at the Joint Research Centre, Ispra Site, Italy, represents a direct contribution from the European Commission to fusion research [2]. ETHEL has been constructed as a multi-purpose facility, essentially experiment independent. This facet will aid in directing the laboratory to new technological goals, as future research needs of the International Thermonuclear Experimental Reactor (ITER) or other fusion devices become invariably modified.

BASIC PROPERTIES AND CHARACTERISTICS OF TRITIUM [3]

Of the three isotopes of hydrogen, protium and deuterium are stable and represent, by far, the vast majority of hydrogen found in nature. Tritium, with a mass number of 3.016, is also a natural isotope, being the result of nuclear reactions induced by cosmic radiation in the upper atmosphere. The calculated natural tritium inventory (atmospheric and oceanic) is several kilograms. Its abundance on earth, however, has been substantially increased during this century owing to research and testing of thermonuclear weapons and, more recently, from nuclear power plants.

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[1]Within fusion research H, D and T are usually employed to denote protium, deuterium and tritium respectively with Q globally representing all three hydrogen isotopes. Likewise, HT often describes elemental tritium with HTO symbolizing tritium oxide.

Tritium decays with a half-life of 12.323 years according to the equation:

$$^{3}T \longrightarrow {}^{3}He + \beta^{-} \qquad (2)$$

The maximum decay energy is about 18.5 keV with a mean of 5.7 keV, values significantly lower than other radioisotopes. Owing to this low energy, the range of the β particle is also extremely limited reaching a maximum of about 6 mm in air with a mean value of 0.5 mm. The corresponding ranges for a 18.5 keV β particle in water, aluminium and stainless steel are about 6 μm, 2 μm and 1 μm respectively. The relatively short half-life and low molecular weight of tritium leads to the radioisotope having an appreciable specific activity of 356 $TBq \cdot g^{-1}$ ($\approx$10 $kCi \cdot g^{-1}$) while its activity density is 95.4 $PBq \cdot m^{-3}$ at STP.

The toxicity of tritium is basically dependent on the form of the radioisotope. As an elemental gas, the derived air concentration (DAC) is $\approx$20 $GBq \cdot m^{-3}$ (56 $\mu g \cdot m^{-3}$), a relatively high value calculated solely from anticipated lung doses the mean tritium β particle range being small compared to the depth of basal skin. However, when tritium is in an oxide form, it can rapidly distribute itself in the body via the lungs or skin such that the biological half-life is only about 10 days. As a result, the DAC for tritium oxide is 800 $kBq \cdot m^{-3}$ (2.2 $ng \cdot m^{-3}$) suggesting that the oxide form is 25000 times more radiotoxic than the elemental gas.

Apart from its radioactivity, tritium behaves physically and chemically in a similar manner to protium and deuterium. Certain physico-chemical effects, however, may be modified by either the ionising β particles or the presence of decay ^{3}He. Indeed, the tritium β particle carries sufficient energy to break chemical bonds leading to radiolytically catalysed interactions when tritium is present. Polymers, especially when halogenated, suffer significant radiolytic damage with relatively low tritium dose rates. Moreover, even glasses, ceramics and metals can have their bulk or surface structures modified when exposed to large quantities of tritium over long durations.

Like protium, tritium dissolves in most materials, an attribute which can lead to significant tritium inventories. In polymers, glasses and ceramics. Elemental tritium dissolves as molecules, while the dissolution process is atomic for metals. There is no single trend between solubility and temperature as the former may increase, remain relatively stable or decrease with temperature for a given material depending on whether the dissolution process is exothermic or endothermic. The velocities with which H and D move through a material, *ie* its diffusivity, are well known for a variety of materials. As expected, diffusion coefficients rise with increasing temperature although, as with the solubility, they are significant differences between materials. Classical theory suggests that the diffusivity of tritium should be about 60% that of protium. The permeability of a diffusing species, *ie* a measure of how rapidly the species will escape from a containment, is the product of the solubility and diffusivity at high driving pressures. For elemental tritium, the permeability always rises with temperature whatever the material although, between materials, the rate of increase varies enormously.

Unfortunately, while the above discussion has generally focused on elemental hydrogen, the permeation of other hydrogen bearing molecules must not be forgotten. This is especially true for HTO which can have a permeability through polymers well in excess of HT.

BASIC LABORATORY DESIGN

ETHEL is a completely new tritium facility foreseen to be licensed for 37 PBq (1 MCi or 100 g) of tritium. The two storey building occupies about 13000 m^3. Much of the laboratory's ground floor is associated with two experimental tritium research areas: the Laboratory for Exploratory Research (LER) and the Hall for Process Development (HPD), Fig. 1. The former is envisaged for low inventory (<37 TBq) experiments, especially those of a complex nature. This is in contrast to the HPD which may contain up to 7.4 PBq in a single circuit.

The foremost operational safety principle applied to ETHEL is the provision of a cascade of barriers and associated pressure differentials to ensure a high reliability of contamination protection, when large quantities of tritium are handled. With few exceptions, tritium, in either its elemental or oxide forms, is held within two distinct containments. Beyond these, the principal protection for operators and the public for reducing potential dose levels in the unlikely occurrence of a significant tritium discharge, is provided by the building's Heating & Ventilation

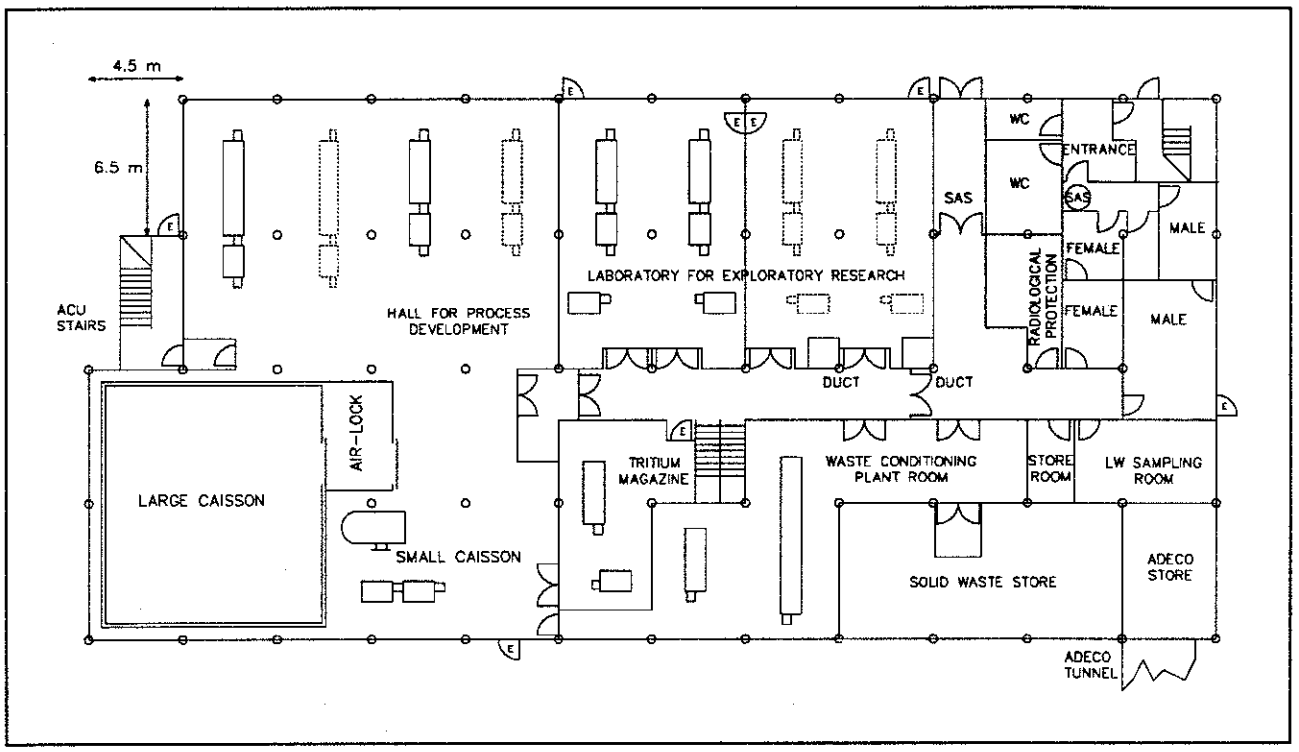

Fig. 1. ETHEL's Ground Floor Layout.

(H&V) System and the various Gaseous Detritiation System (GDS) units, Fig. 2. The latter primarily serve to maintain the tritium concentration in glove-boxes sufficiently low to preserve doses due to chronic releases at fractions of the regulatory levels. Overall, the aim is to restrict air-borne tritium stack releases to <3.7 $TBq \cdot a^{-1}$.

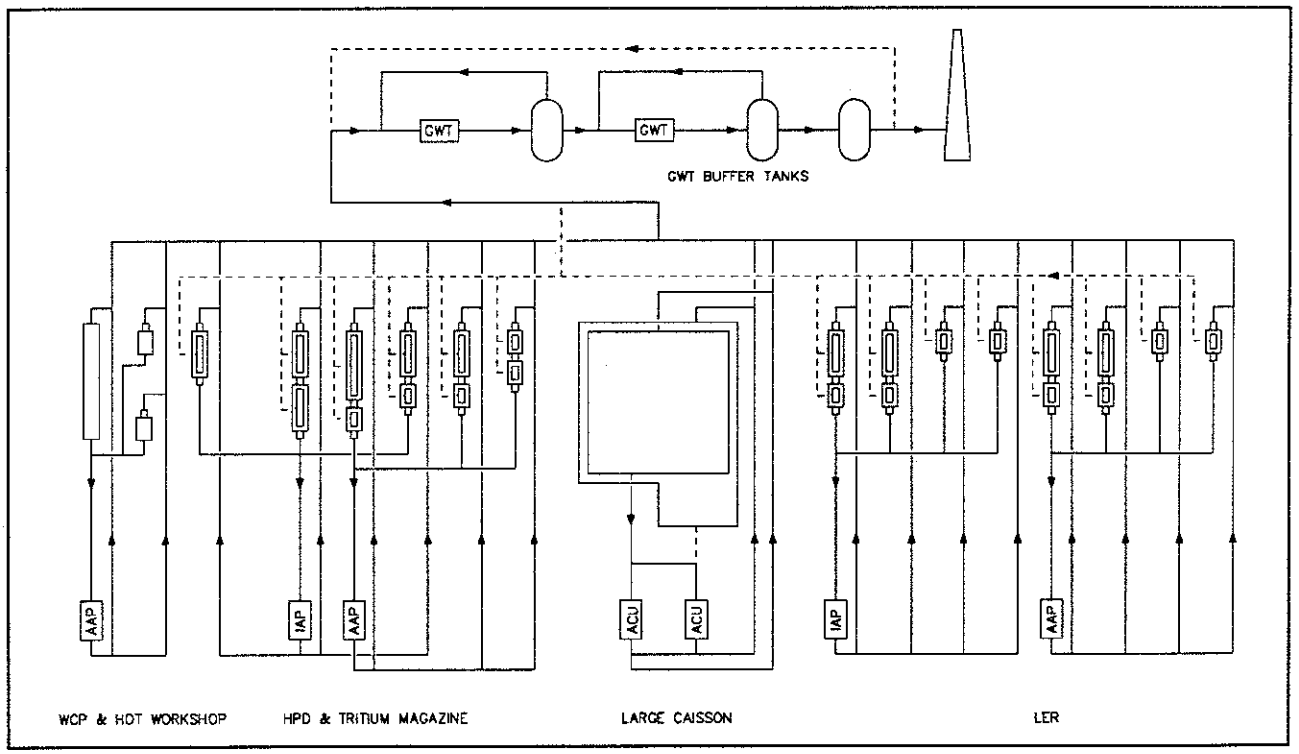

Fig. 2. Overall Gaseous Detritiation System Architecture.

Primary containments

The primary containments, *eg* the process plants and experimental circuits, represent the first barrier. Given the mobility of elemental tritium, such containments must have a high standard of overall leak tightness, typically 10^{-5} - 10^{-7} $Pa \cdot m^3 \cdot s^{-1}$ with individual components specified at $\leq 10^{-9}$ $Pa \cdot m^3 \cdot s^{-1}$, all test differential pressures being one atmosphere. An exception to this rule is the 350 m^3 Large Caisson, a double skinned stainless steel chamber which has primary and secondary leak rate specifications respectively of ≤ 0.01 vol.%$\cdot h^{-1}$ and ≤ 0.05 vol.%$\cdot h^{-1}$ at 1 kPa ΔP.

Checking the overall leak tightness of primary circuits, especially when contaminated, is problematic. For most circuits, an integral turbo-molecular or drag pump system must be provided to render possible standard vacuum leak test techniques. As can be imagined, such methods are not practical for the Large Caisson given the dimensions and leak rate specifications of the containment. Instead, the rate of concentration increase of chemical tracers is employed. More specifically, to measure the primary containment leak rate, N_2O is injected into the interspace between the two metal skins and a differential pressure held between the interspace and inner containment, Fig. 3 [4, 5].

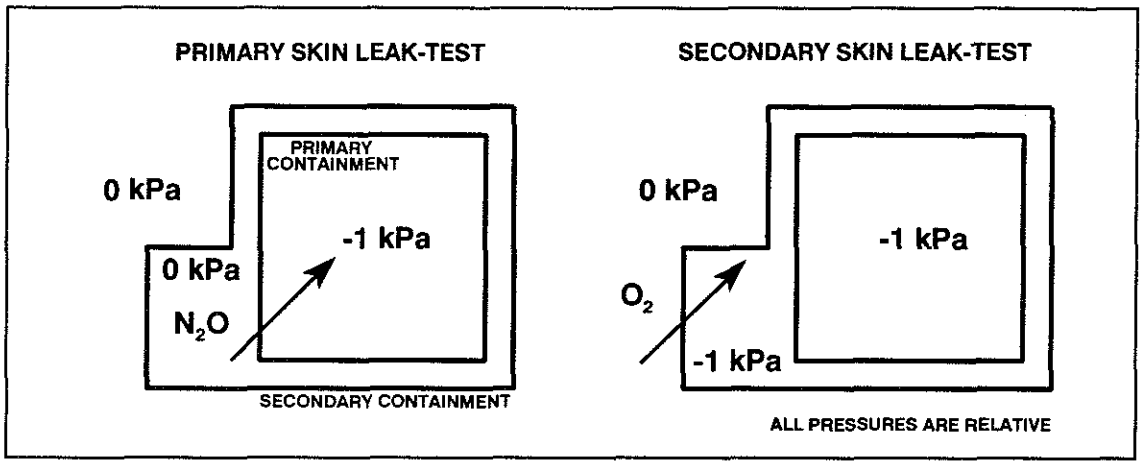

Fig. 3. Large Caisson Leak Testing.

For obvious reasons, this approach cannot be repeated for testing the outer skin. Instead, the outer skin leak rate can be ascertained from monitoring the increase in interspace oxygen concentration using room air as the tracer.

Obviously, any material coming into direct contact with tritium must be adequate for the intended service conditions. On occasions, however, materials which appear totally adequate from a compatibility viewpoint, may be unacceptable for other reasons, one of which is often permeability. This is especially true when high operating temperatures are encountered and where, for such conditions, use may be made of exotic materials, *eg* tungsten, gold. Fig. 4 shows how the permeation of tritium varies with temperature for a variety of materials employed in ETHEL experiments [6].

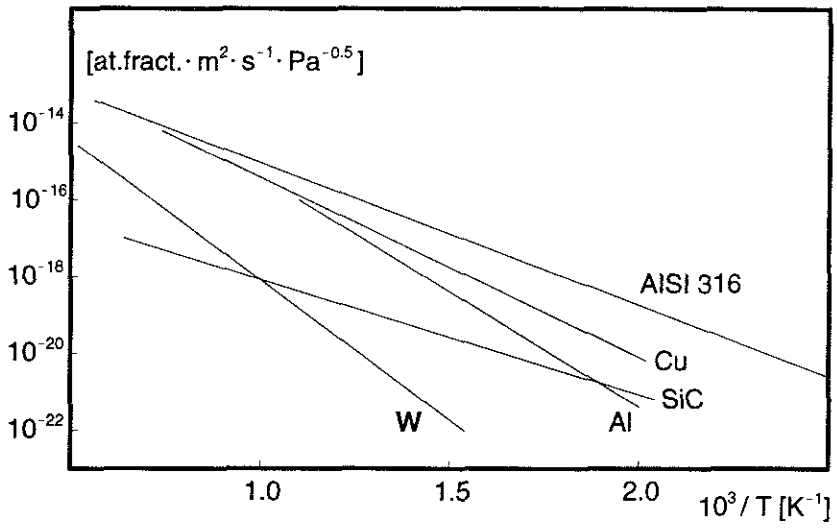

Fig. 4. Variation of Permeability with Temperature.

Experiment (Secondary) Containments and Associated Gaseous Detritiation Units

Since there are no major shielding requirements, tritium experiments are performed in glove-boxes. Although not ideal (nominal leak rate ≤0.05 vol.%·h^{-1} at 1 kPa ΔP), the glove-boxes offer adequate protection in view of their associated gaseous detritiation units. However, like primary containments, the periodic verification of glove-box leak rates can be troublesome, especially when secondary containments of several cubic metres are encountered or when significant quantities of tritium are stored within. In such instances, the application of any leak test in which the temperature is an important parameter is questionable. To overcome this, ETHEL is considering to employ a similar oxygen test to that already used for the Large Caisson.

It has already been noted that the permeation of tritium through materials of diverse types, *eg* polymers, metals, ceramics, can vary by several orders of magnitude. In a similar manner, permeation rates among different polymers can cover a large spectrum. Unfortunately, the problem is further compounded since many polymers which display a low permeation rate for HT are poor at hindering HTO and *vice versa*. For this reason, the gloves employed on secondary containments are made from a laminated polymer of neoprene and butyl rubber; the former hindering HT and the latter HTO [7].

The glove-box atmospheres are either dry air or argon. Depending on the atmosphere, one of two types of GDS unit is used: Air Atmosphere Purification (AAP) units for air and Inert Atmosphere Purification (IAP) units for inert environments. Both AAPs and IAPs function by oxidising hydrogen isotopes on a precious metal catalyst bed and trapping any water on molecular sieve beds. Design targets of <37 MBq·m^{-3} in the glove-boxes and <2 ppm H_2O at the exit of the sieve beds have been established for normal conditions [8]. The former value represents about 10^{-7} g of tritium per cubic metre of glove-box atmosphere or, in classical terminology, <0.1 ppb.wt. The oxidation catalyst cannot be expected to perform efficiently at such low concentrations. Therefore, to enhance the oxidation process, the elemental tritium in the process stream is isotopically swamped by the addition of H_2, Fig. 5. A similar argument can be made for the molecular sieve beds in which case H_2O vapour is mixed with the oxidised process gas.

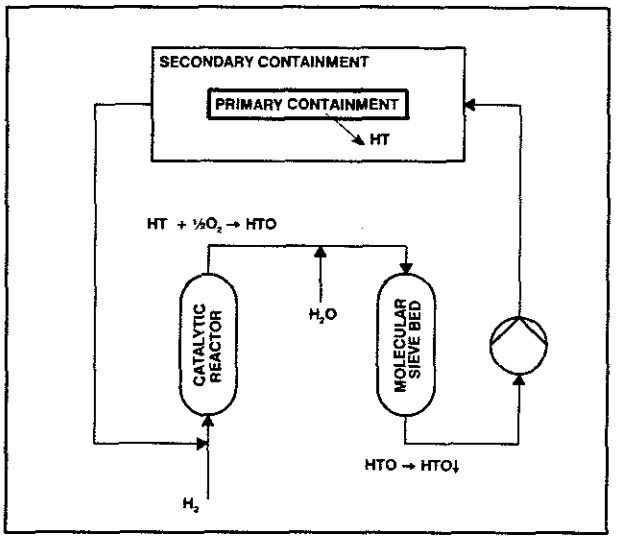

Fig. 5. Gaseous Detritiation Unit Flow Diagram.

One drawback of molecular sieves in tritium technology is memory effect which can arise for two reasons: HT and/or HTO isotopically exchanging with the sieve bed bulk material and residual HTO after sieve bed regeneration. For instance, an often used molecular sieve, type 5A, has a residual water content in excess of 1% at a relatively high regeneration temperature of 575 K. Obviously, it is possible to reduce this quantity of water by regenerating at a higher temperature but this is expensive and can, in the long term, lead to mechanical breakdown of the bulk material and associated dust problems. Presently, there is no satisfactory solution to these problems. Hence, much research is being conducted to develop suitable sieve bed substrates which mirror the basic drying performance of conventional materials without their aforementioned disadvantages.

Tertiary containment

The function of a kind of tertiary containment is offered by the building in combination with the negative differential pressure created by the H&V system with respect to the generic public against exposure by ground level releases. For operational purposes, the building has been divided into three radiological zones: clear zone, low risk controlled area and medium risk controlled area, in which three bands of increasing pressure differentials are maintained. In the event of a discharge of tritium within the laboratory, the H&V system is programmed to respond to an *area tritium high* level indication by increasing the flowrate in that area, *ie* to discharge any releases as fast as possible to the environment. This approach is justified for ETHEL given the limited tritium inventory of the laboratory (in respect to a fusion device), the negligible increase in dose to the critical group of the population and the anticipated reduction in operator exposure due to the enhanced dilution of the tritium concentration and the reduction of its residence time, by which its absorption on and successive slow release from the surfaces in the concerned area will be minimised.

Storage and Control of Tritium

Within the Magazine, tritium is stored as solid uranium tritide on six fixed getter beds, each with a capacity of >9.3 PBq at 50% saturation. Storage as a tritide rather than in the gaseous form is preferred because of the inherent safety and compactness of getters. Unfortunately, uranium getters have two major drawbacks. The first is the pronounced pyrophoricity of the metal especially when in a powder form. Consequently, the Tritium Magazine glove-box atmosphere is argon. The fire extinguishant for the secondary containment is also argon since uranium readily reacts with CO_2. The second disadvantage of uranium is its inherent ability to irreversibly trap a small, but significant quantity of tritium which can only be removed by swamping with other hydrogen isotopes. Without direct measurements, the uncertainties associated with the heel can seriously influence tritium inventory calculations, because the best present day estimates of the heel are accurate to only ±70%.

Unlike fissile materials which are often encountered in well defined physical and chemical forms with a reasonable radiation fingerprint, the distribution of protium in materials, the extreme mobility of hydrogen isotopes and ease of contamination and, finally, the feeble, non-penetrating radiation of tritium present enormous complications for assessing tritium inventories, Table 1. These difficulties are especially evident when tritium is held up in process lines (absorbed on surfaces or permeated into bulk materials) or contained in solid wastes. Moreover, for solid wastes, the only currently known precise method of determining the tritium inventory is by total destructive analysis, a technique which is rarely applicable.

Table 1. Basic Differences between Tritium and Fissile Materials [9].

	TRITIUM	FISSILE MATERIALS (^{239}Pu - ^{235}U)
Typical form	Gas, liquid or solid	Liquid or solid
Chemical mobility	Extremely mobile	Practically immobile
Radiotoxicity	HT very low, HTO low	^{239}Pu high, ^{235}U moderate
Half-life	12.323 a	^{239}Pu $2.4 \cdot 10^4$ a ^{235}U $7.1 \cdot 10^8$ a
Radiation footprint	β^- (max. energy 18.5 keV)	^{239}Pu ($\alpha \approx 5.1$ MeV) ^{239}Pu ($\gamma \approx 0.04$ - 0.77 MeV) ^{235}U ($\alpha \approx 4.4$ MeV) ^{235}U ($\gamma \approx 0.14$ - 0.20 MeV)
Typical inventories	mg - g	g - kg
Measurement	PVT - concentration Calorimetry Weight - concentration	Passive NDA Active NDA Calorimetry - concentration Weight - concentration

Whereas there is a large variety of techniques for measuring quantities of fissile materials, the detection, let alone precise quantification of freely measurable tritium is more problematic. Also, it is worth recording that inventory measurements in facilities such as ETHEL are often in the milligram range with gram quantities of tritium being the exception. This contrasts sharply with fissile material controls where gram and kilogram quantities are routinely encountered. However, the uncertainties associated with tritium held up either permanently or temporarily in process plant, waste, *etc*, must not be underrated.

Of the three basic quantification methods for determining the tritium inventory, the ideal gas equation is widely applied. Apart from potential precision, the main advantage of this approach is its extremely wide dynamic range which is basically a function of the pressure measurement. There is a number of handicaps, however, such as possible difficulties in measuring a representative temperature or accurately determining the tritium concentration even though a variety of techniques are available, *eg* gas chromatography, mass spectrometry, Raman spectroscopy. Gas chromatography is a method of analysis in which a flow of solvent, *eg* carrier gas, promotes the separation of substances such as hydrogen isotopes from a narrow initial zone in a porous sorptive medium. Mass spectrometry employs the fact that ions formed in an ion source can be separated by appropriate magnetic or electrical fields and subsequently quantified. Finally, Raman spectrometry is based on measuring the intensity of Raman scattering produced when light impinges on polyatomic molecules.

Whereas, the PVT-c gas technique requires a knowledge of the tritium gas purity and, hence, an analytical measurement to determine the presence, if any, of likely contaminants such as other hydrogen isotopes, helium and argon, calorimetry can often be employed as a non-destructive assay (NDA) technique for the absolute determination of tritium inventories. Moreover, by avoiding gas samples, both operator involvement is reduced and tritiated waste from the analytical technique eliminated. Unfortunately, one fundamental drawback of calorimeters is their rather limited dynamic range, typically <1000.

The weight measurement technique is self-explanatory. However, the weighing of tritium is not always readily applicable owing to the small mass of routine tritium inventories and typical weight of tritium containers; pressure vessels or getters including any associated pots usually weigh several kilograms. For instance, the weight of a portable getter used in ETHEL will primarily consist of the blank getter and containment pot with tritium (readily available or as a heel) and ^{3}He in the getter and Ar in the containment pot interspace representing the balance, Fig. 6. Consequently, like calorimetry, this large background effectively reduces the dynamic range of weight measurement techniques for tritium [10].

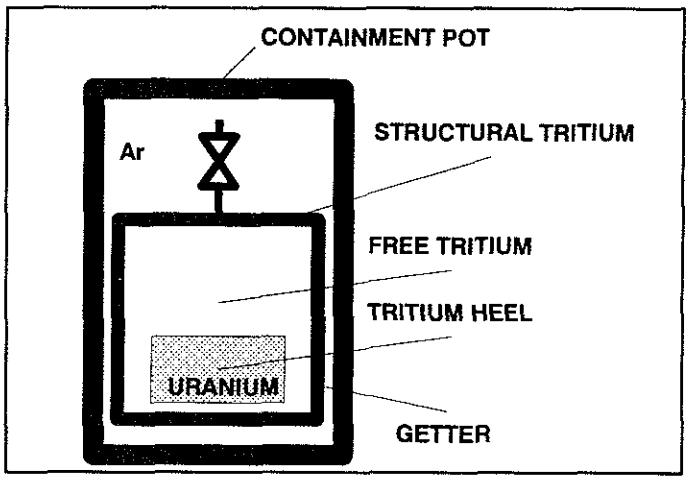

Fig. 6. Typical Tritium Distribution in a Portable Getter.

TRITIATED WASTES

The Ispra Site is not served by a central waste treatment station capable of handling heavily tritiated wastes. Hence, ETHEL is equipped with a Waste Conditioning Plant (WCP), designed to accept tritiated wastes, both liquid and solid, and to perform routine operations on such materials under safe working conditions [11]. The plant, the majority of which is located inside a 14 m^3 glove-box suite, allows a variety of operations including waste storage and sorting, vacuum re-packaging of bagged solid wastes, liquid waste solidification by dry cement hydration or sorption on vermiculite, reversible aqueous condensate immobilization by sorption on molecular sieves, solid waste encapsulation by cement grout and drumming under tritium containment conditions. Such operations are necessary, since ETHEL is not allowed to store, on a long-term basis, free tritiated liquids nor loose tritiated solids.

A variety of problems such as inventory control, waste packaging materials and decay tritium over-pressures, can be encountered when conditioning tritiated wastes.

The maximum activity of tritium which can be irreversibly immobilised is restricted to about 1.85 GBq (50 mCi) per kg of conditioned solid waste as required by the Italian Regulatory Authorities [12]. Likewise, a knowledge of the tritium inventory in conditioned waste is a prerequisite for fully implementing the laboratory's tritium control procedures. Unfortunately, the direct determination of the tritium inventory in solid wastes is impossible. Instead, indirect approaches must be applied, although presently these only lead to qualitatively providing the tritium content in medium and high level solid wastes. Apart from the history of the waste, indirect techniques depend on the measurement of surface activity and out-gassing rates though the latter may be influenced by temperature, humidity, surface conditions, *etc.* Also, the bulk material of tritiated solid waste is often

not homogeneous, *eg* valves, pumps, instrumentation. As a consequence, the development of improved techniques for quantifying tritium inventories in solid wastes is a fundamental objective of operating ETHEL.

The packaging of soft wastes and drum liners represents additional barriers for inhibiting the dispersion of tritium to the building's ventilation system and, hence, environment. Historically, the tritium community has employed high density polyethylene (HDPE) as flexible bagging or packaging as this polymer represents a good permeation barrier to HTO, the much more radiotoxic form of tritium. In addition, HDPE displays excellent radiation resistance, is readily heat sealable, inexpensive and, from a quality viewpoint, easy to control. The major drawback of HDPE is its inability to effectively hinder the permeation of HT which may subsequently convert to HTO outside of the waste package.

While not refuting the benefits of HDPE, other packaging materials can offer significant improvements in reducing the permeation of both HT and HTO. The food industry has vast experience in packaging materials owing to the need to stop the ingress of O_2 and H_2O into prepared packets. While that industry is still a large user of polyethylene, other polymers such as polyesters are frequently employed. In addition, as modern manufacturing techniques enable multi-layer films to be easily fabricated, single polymer films have often been displaced by composite packagings. However, of even more interest to tritiated waste management is the possible incorporation of aluminium in packaging materials. Indeed, the potential benefits are so attractive that research in ETHEL is directed to identify and test composite packaging materials for tritiated waste conditioning with initial focus being given to a 4 core film, Fig. 7. Even under adverse operating conditions, this packaging will lead to an order of magnitude decrease in HTO permeation rates compared with the equivalent thickness of HDPE while the corresponding HT permeation rate should be reduced by >1000 [13]. Moreover, such a film will still maintain the heat sealing properties of HDPE while showing improved mechanical resistance against tears, perforations and abrasion.

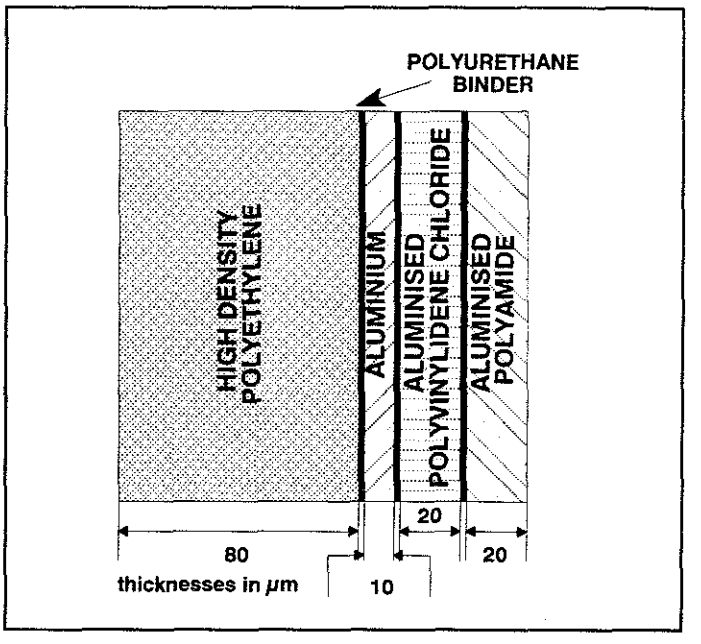

Fig. 7. Improved Packaging for Tritiated Wastes.

Not all waste in ETHEL is irreversibly conditioned. Small volumes of highly tritiated water are solidified by sorption on zeolites into suitable leak tight aluminium bottles which are then drummed for retrievable storage. Each bottle must be designed to withstand a high pressure increase due to the ^3He production from tritium decay and radiolysis of sorbed water. Quantitatively, an internal pressure of 2 MPa is anticipated in the 5 dm^3 after storing 56 TBq for five years [11].

After waste conditioning, each drum is tested for out-gassing and surface contamination, prior to being sent to the ETHEL's Solid Waste Store (SWS), a bunker within the laboratory. The SWS has been designed to allow the controlled on-site storage of all packaged wastes produced in ETHEL such that any gaseous tritium releases are measured before being discharged to the stack. This release to the 80 m stack is an implicit requirement of the site license since losses at ground level would lead to a forty fold reduction in the allowable laboratory discharges, *ie* 40 instead of 1600 Ci per year.

HEALTH PHYSICS

The radiation protection principles applied to the design and construction of the facility are aimed at effectively optimising the protection of both workers and the population. The objective is to keep irradiation doses and discharges well below the regulatory limits. The overall collective effective dose equivalent was taken to guide the designers in the elaboration of the safety case proposed to the Regulatory Authority. The annual individual dose for the most exposed worker during the normal operation and maintenance of the laboratory is foreseen to be significantly lower than 1 mSv. The goal for the collective effective dose equivalent is 10 mSv. Ultimately, the

intention is to demonstrate the possibility of operating facilities with high tritium inventories such as ETHEL with an effective dose equivalent for ≤ 10 $\mu Sv \cdot a^{-1}$ to the critical group of the population even in a highly populated area.

Non-discriminating ionisation chamber monitors are employed as the standard tritium detection device within ETHEL. Obviously, this implies that no distinction is made between HT and HTO, an approach which provides a considerable safety margin. The standard monitor employed in the laboratory is based on a 2 dm^3 ionisation chamber which, together with the associated electronics, has a dynamic range $\geq 10^7$ and a lower sensitivity limit of 37 $kBq \cdot m^{-3}$. A monitor is located in each individual glove-box near the exhaust duct leading to the associated GDS unit. The monitor is of an open wall design (perforated type), *ie* gas may freely flow through the chamber without the need of a dedicated pumping system. This concept has advantages of increased overall system reliability and a faster response time in case of an accidental release inside a glove-box.

The position of monitors surveilling the tertiary containment is more varied. For glove-boxes, two problems must be recognised. Firstly, the need to obtain a representative sample of the glove-box working face and, secondly, to rapidly obtain such a sample. Although no perfect solution has been identified, the present idea is that one flow through ionisation chamber provided with sampling inlet-outlet orifices (closed type) is located above each secondary containment, samples being collected through a manifold and pumped through the chamber.

The main ETHEL exhaust duct is provided with two monitors. One is of the closed type described above and is employed for real time surveillance. The second is a discriminating monitor based on two pairs of bubblers and an oxidation catalyst offering a significantly increased sensitivity to the ionisation chambers. Samples from this system are removed at least every 10 days to verify the total tritium discharges and identify the elemental/oxide fractions. Besides the above, a near real time discriminating monitor capable of distinguishing between HT and HTO in room air is under development in ETHEL. The monitor uses a nafion membrane to separate the two species and presently provides quantitative discrimination in a few minutes, Fig. 8, [14].

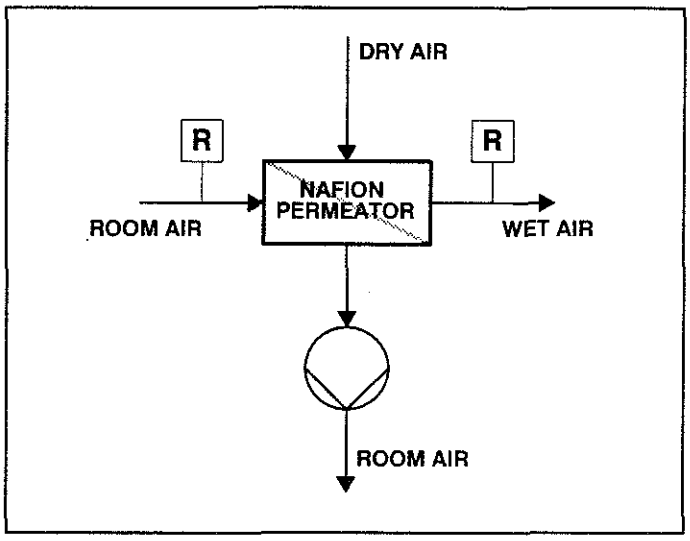

Fig. 8. Flow Diagram of Discriminating Monitor.

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