
Advanced Tritium Recovery System

Introduction

As part of the U.S. Inertial Confinement Fusion Program, LLE plans to implode deuterium–tritium (DT)–charged microspheres. These microspheres are filled with high-pressure deuterium and tritium gas under cryogenic conditions. The spheres are then stored and transferred as needed to the target chamber (located in an adjacent room) under vacuum and at temperatures near 20 K. LLE is licensed to handle up to 10,000 Ci of tritium, but site release target is less than 1 Ci per year.

LLE is currently upgrading the systems used to produce, transfer, store, and implode these targets. As part of this upgrade, extensive efforts are being made to ensure staff safety and to limit the radioactive effluent that can be released to the environment. Two independent tritium recovery systems (TRS), currently under construction, will capture any tritium that comes from the various sources. One system will serve Room 157, which houses the production, characterization, and storage of targets, and the other system will serve the target chamber, where the targets are imploded. The exhaust emissions from the process streams, from the gloveboxes that house the streams, and from equipment outgassing will have to be collected and treated to remove any tritium present. This article provides an overview of the requirements and describes the technologies that were considered for the decontamination of the process streams and glovebox-cleanup systems for Room 157.

TRS Design Considerations

1. General Constraints and Considerations

To maximize personnel safety and minimize site emission, the following factors were considered as part of the decontamination systems' design:

- (a) Critical components and large inventories have secondary or tertiary containment.
- (b) No single component failure results in a release of tritium to the room or to the stack.

- (c) Systems containing tritium are designed for high integrity, having all stainless components, with either welded connections or leak-tight (VCR) fittings and a minimum of elastomer seals.
- (d) Complex operations are typically automated or, if manual, are covered by detailed procedures.
- (e) Protective systems that are poised are designed to be tested regularly to ensure their operability.
- (f) Active components are designed to allow easy maintenance.

In addition to the design considerations, the operating staff is technically knowledgeable and extensively trained in handling tritium.

While the system has been designed to limit the likelihood of accidental emissions to a very low probability, additional features are included to minimize chronic releases. These can be summarized as follows:

- (a) Since the radiotoxicity of HTO is about 10^4 times greater than that of HT, every effort is made to avoid oxidizing any HT present to HTO.
- (b) Mixing of exhaust streams having different gases or tritium concentrations is minimized to limit the quantities of effluent to be decontaminated.
- (c) All exhaust streams are monitored and controlled.
- (d) All accessible areas are well ventilated to avoid potential tritium buildup due to outgassing from contaminated equipment.

2. Room 157 Functional Requirements

To achieve the above objectives, the Room 157 effluent gases are divided into the following four categories:

- DT high-pressure-system (DTHPS) secondary containment (SC) cleanup
- Glovebox (GB) cleanup (low Y, inert)
- Helium vacuum exhaust cleanup [high temperature (high T), inert]
- Air vacuum exhaust cleanup [low temperature (low T), air]

The relationships between the streams and the subsystems that release these streams are provided in Fig. 89.23.

This sorting was done in order to select the best technology for each type of stream and, as a result, achieve the best performance (i.e., lowest emissions) at a reasonable cost.

The DTHPS handles pure DT under high pressure, so it has its own secondary containment, which, in turn, has its own tritium-scavenging system to collect any DT that escapes from the DTHPS. This DTHPS secondary containment and its DT scavenging system are housed inside the DTHPS glovebox. For these reasons, it is impractical to combine its effluent with any of the other systems.

The second category consists of the gloveboxes that have helium atmospheres. Three process trains exist: one for the DTHPS glovebox atmosphere, one for the FTS (fill transfer station) and TFS (tritium fill station) glovebox atmospheres,

and one that acts as a backup to either. Each train consists of a circulating pump, a drier, and, finally, an HT removal bed. Interconnections are provided to allow various combinations of pumps, driers, and HT removal beds. The driers serve primarily to remove any moisture that permeates into the gloveboxes in order to avoid excess loading of the tritium scavengers, but the driers will also remove any HTO that may be present.

The rationale for further splitting the glovebox-cleanup function is that the DTHPS has its own internal containment and the DTHPS glovebox represents a tertiary containment. It will normally have no or very low levels of tritium, so it will require essentially no cleanup on an ongoing basis. The FTS and TFS gloveboxes have comparable levels of tritium activity, so mixing these will have no significant effect on cleanup. Another factor is the potential for dilution. Since the effectiveness of cleanup is to some extent driven by the initial concentration of tritium, mixing essentially clean streams with contaminated streams actually decreases the tritium concentration in the contaminated stream, resulting in reduced capture efficiencies in some devices.

The third category consists of the vacuum systems that service the helium processes. Again, a further subdivision of this group separates the “clean” sources, specifically the FTS base, dome and cooling loop, permeation cell seal, stalk

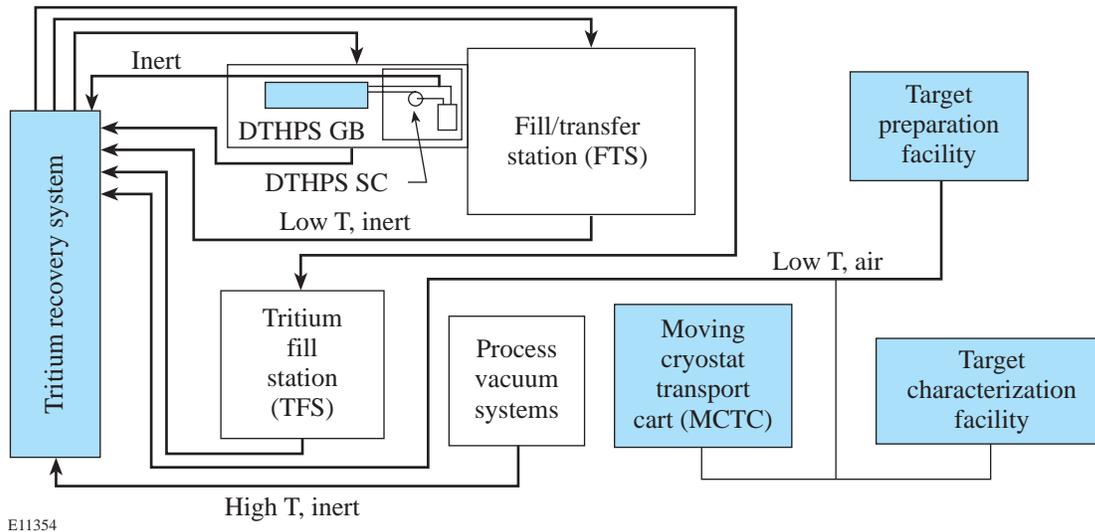


Figure 89.23
The Room 157 Tritium Recovery System must deal with air and inert gas streams from a variety of sources.

aligner, and borescope, from potentially contaminated sources. Using a monitor to confirm the absence of tritium, the exhaust is routed directly to the stack. If tritium activity is detected, the stream is combined with the other helium vacuum exhaust streams for treatment.

All other helium vacuum exhaust streams are combined into a collection tank. A pump circulates the gas through a drier to remove any HTO, a Ni catalyst bed cracks any organically bound tritium, and finally a scavenger bed removes the product HTO. The subsystem pressure is capped by a relief valve that discharges to a second tank, which can also recirculate any effluent through a second scavenger. The gas in this second loop should be essentially tritium free and should be releasable to the environment. Any discharged gas is nevertheless monitored, and, if it requires further decontamination, it can be routed to the air vacuum exhaust for additional processing.

The final category comprises air vacuum exhaust streams. These streams are not subdivided since they are all normally active, so they will require a routine processing. The stream is initially passed through a drier bed to remove any tritium that might be present as HTO. This may be sufficient to allow the exhaust to be released to the stack, so the stream is monitored at this point and, if possible, sent to stack. If the level of tritium activity is still above the release criteria, the stream is routed to a catalytic oxidizer to convert any tritium that may be present as HT or organically bound tritium (OBT) to HTO. This effluent is then passed through a condenser and another drier bed before going to stack. The final effluent is once more monitored for compliance purposes.

The catalytic reactor most suitable for the LLE application is palladium-coated alumina operated at high temperature (~450°C) and sized to give a residence time of about 4 s. The reactor is followed by a chiller to cool the exhaust before it goes into a drier bed. The catalytic reactor, chiller, and drier beds used in this application are designed for high flow and low resistance to minimize the need for additional pumps to move the gas through the detritiation loops.

All the tritium removal beds have provision for *in-situ* regeneration to eliminate the need for routine bed removal. The regeneration of the HT scavenging beds is accomplished by heating the bed and using a helium carrier flow to accumulate the released HT on a cryogenic molecular sieve before it is transferred to a transportable U-bed for eventual removal from the system. Thus, any captured HT will remain in the elemental form throughout the entire tritium cycle.

The drier beds are also heated for regeneration, releasing the HTO, which is carried by an N₂ gas carrier to a chiller followed by a condenser to remove the bulk of the moisture. The gas then flows to a polishing drier bed, is monitored, and finally released to stack. The recovered moisture, including HTO, is collected in liquid form for interim storage, pending final disposal. Regeneration of driers is by reverse flow to counteract HTO tailing in the drier beds. Hot, dry nitrogen gas is used as a carrier to achieve dew points below -100°C. This allows an extended operating cycle while still maintaining the low outlet tritium concentration needed to meet emission targets.

Tritium Removal Technologies

A number of viable options are available for removing HT from inert gas streams, and given the number of helium-based streams at LLE, every effort was made to use this capability to minimize the production of HTO. The following section examines the technologies that were applicable to the LLE Tritium Removal System.

1. Uranium Beds

Uranium reacts exothermically with hydrogen at room temperature, forming UH₃. Uranium has a high storage capacity for hydrogen (up to 300 scc/g U) and uniform reaction kinetics over a very large H/U ratio. The decontamination factor varies depending on flow and tritium concentration, but a practical minimum effluent concentration of ≈ 2 mCi/m³ is achievable.¹

The hydriding reaction is reversible, and by heating the hydride to ~400°C, the hydrogen is released, restoring the bed to its original state. Varying the amount of heat applied can control the hydrogen release rate. However, uranium reacts not only with hydrogen, but also reacts strongly with oxygen and somewhat with nitrogen at room temperatures. Both the oxidation and nitriding are exothermic and become more vigorous at elevated temperatures. When purged with air, U-beds will release stored tritium.

For this reason, uranium can be used only for tritium removal from inert gas streams. It will remove large amounts of tritium with high efficiency and will do so for considerable carrier gas flow rates. Its affinity for oxygen and, to some extent, other gases serves to purify the tritium since only the hydrogen isotopes are released when uranium is heated, although at the cost of irreversibly reacting with the uranium. U-beds have been extensively used, and a very large experience base exists to support this application.

Uranium is not recommended for inert glovebox atmosphere cleanup since the practical decontamination level of $\sim 2 \text{ mCi/m}^3$ is higher than desired. It is best used where large quantities of tritium must be removed and where the effluent will not be released to stack.

2. Zirconium-Iron (ZrFe) Beds

The ZrFe alloy known as ST-198 can also scavenge tritium from inert gas streams since it forms a stable tritide that can be reversibly decomposed.² It has a rather modest storage capacity for tritium (0.8 scc/g alloy yields a 1 mCi/m^3 vapor present over the alloy) but can decontaminate to $\mu\text{Ci/m}^3$ levels if tritium inventory of the alloy is kept sufficiently low. Although it requires a substantial residence time ($>3 \text{ s}$) to achieve good detritiation, it is an excellent tritium scavenger for glovebox decontamination and for polishing exhaust streams before stacking. Since ST-198 reacts very slowly with nitrogen at room temperature, it can be used to scavenge HT from nitrogen streams.³ It does not react with organics (CH_4), so it will not scavenge tritiated organics from a gas stream unless the stream has been preconditioned with a nickel catalyst to crack the organics to produce HT.^{4,5} While ST-198 can be regenerated by heating, it degrades during this process and consequently has a limited lifetime.

Like uranium, ST-198 reacts with oxygen at room temperature and with nitrogen at elevated ($\geq 450^\circ\text{C}$) temperatures. Both reactions will release stored tritium. ZrFe is best where quantities of hydrogen isotopes are limited, but where the effluent tritium concentration must be suitable for release to the environment.^{6,7}

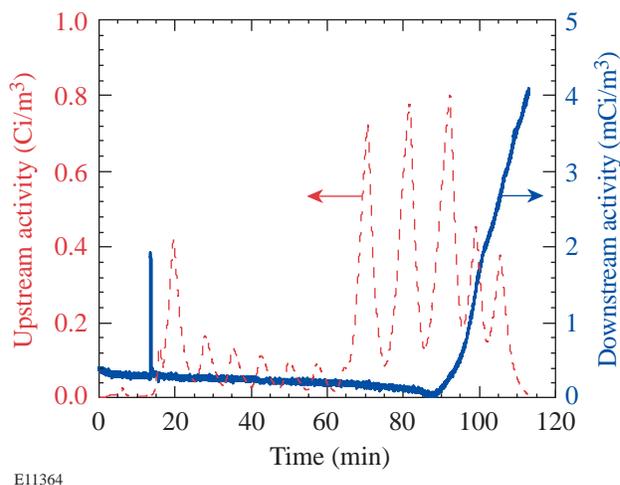
3. Cryogenic Molecular Sieves

Cryogenic molecular sieves can scavenge hydrogen isotopes from a helium carrier by adsorbing the hydrogen molecules. Depending on the partial pressure of hydrogen that is acceptable, molecular sieves operating at liquid nitrogen (LN) temperatures can have a high capacity for hydrogen isotopes. However, the tritium is not firmly bound in the molecular sieve at 77 K, so the tritium residence time in the bed is finite. The residence time is a function of both flow and geometry in that longer beds and lower flows yield a longer time to breakthrough. Warming the molecular sieve slightly above LN temperatures (i.e., to $\sim 100 \text{ K}$) will release the stored hydrogen isotopes. At these temperatures, all other gases will remain firmly adsorbed, thus allowing the recovery of pure-hydrogen

isotopes. These gases can, however, be desorbed by heating the molecular sieve to 350°C , regenerating the molecular sieve to its original condition.

These characteristics make cryosorption an excellent technique to scavenge tritium from helium gas streams. Low exhaust levels of tritium activities can be achieved by limiting the amounts of tritium in the molecular sieve and by monitoring for impending breakthrough. When breakthrough becomes evident, the bed needs to be valved out of service so it can be unloaded. The unloading process proceeds rapidly since the bed needs to be warmed to only 100 K for complete HT unloading.

Figure 89.24 illustrates the efficacy of a cryogenic mole sieve bed to remove HT from a helium stream containing up to 800 mCi/m^3 . During this test, the activity downstream of the bed remains below 0.3 mCi/m^3 until breakthrough occurs.



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Figure 89.24
Transfer of tritium from 800-g ST198 bed to cryogenic mole sieve bed with a 28 sLPM helium carrier until tritium breakthrough is observed.

The chief disadvantage of cryosorption is that it requires liquid nitrogen temperatures to retain the hydrogen isotopes on the molecular sieve. This means that a loss of cryogenics could result in the inadvertent release of hydrogen. This capture technology is not passively safe. In addition, the complexity of cryogenic handling is costly and requires techniques and skills that are not commonplace. An example is the need for regenerative heat exchangers as part of any cryotrap to obtain high utilization of liquid nitrogen inventories.

System Configuration

A comprehensive tritium removal system (presented schematically in Fig. 89.25) has been designed and is being built so that LLE can perform its research program and still meet tritium emission targets. To ensure that this system can provide the desired detritiation performance and also provide the highest practical degree of staff safety, all relevant technologies were extensively reviewed. Based on this review, ZrFe scavenger beds are used to reduce tritium levels to low values, cryogenic molecular sieves are used to concentrate tritium, and uranium flow-through beds are used to capture large amounts of tritium. Nickel catalysts are used to crack organics, Pd on alumina is used to oxidize any tritium compounds in air systems, and 5A molecular sieve driers were selected to remove HTO. The technologies selected for the final design were those judged most suitable based on demonstrated robustness, effectiveness, and real-life operating experience.

ACKNOWLEDGMENT

This work was supported by the U.S. Department of Energy Office of Inertial Confinement Fusion under Cooperative Agreement No. DE-FC03-92SF19460 and the University of Rochester. The support of DOE does not constitute an endorsement by DOE of the views expressed in this article.

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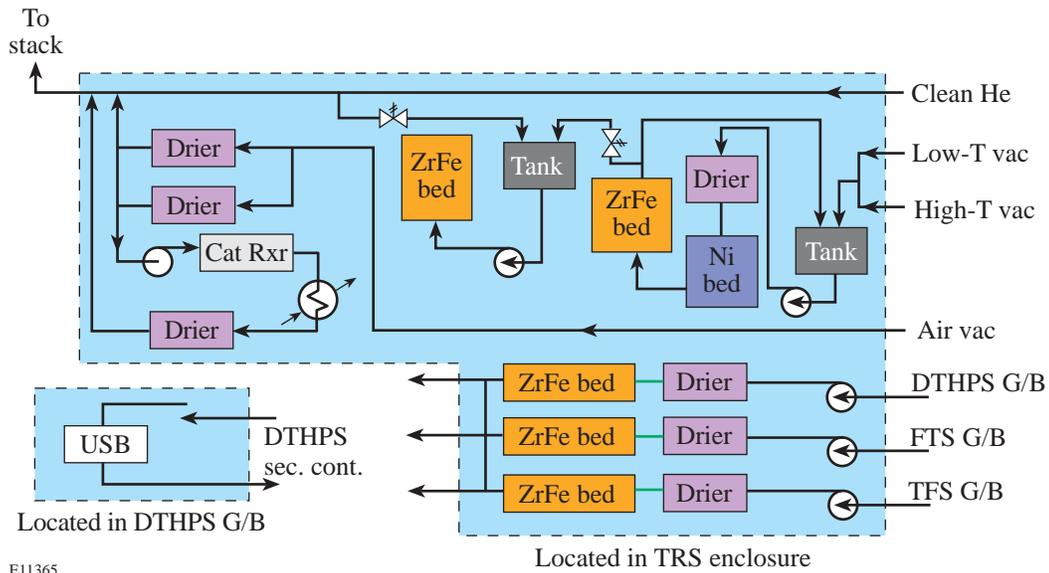


Figure 89.25
Schematic of the Room 157 Tritium Recovery System.

