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Glovebox Stripper System Tritium Capture Efficiency - Literature Review

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EXECUTIVE SUMMARY

Glovebox Stripper Systems (GBSS) are intended to minimize tritium emissions from glovebox confinement systems in Tritium facilities. A question was raised to determine if an assumed 99% stripping (decontamination) efficiency in the design of a GBBS was appropriate. A literature review showed the stated 99% tritium capture efficiency used for design of the GBSS is reasonable. Four scenarios were indicated for GBSSs. These include release with a single or dual stage setup which utilizes either single-pass or recirculation for stripping purposes. Examples of single-pass as well as recirculation stripper systems are presented and reviewed in this document.

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LIST OF ABBREVIATIONS

DF	Decontamination Factor
GBBS	Glovebox Stripper System
INL	Idaho National Laboratory
OSTI	United States Department of Energy, Office of Scientific and Technical Information
ppm	Parts per million
SHINE	SHINE Medical Technologies ™
SRNL	Savannah River National Laboratory
STAR	Safety and Tritium Applied Research (Facility)
TRL	Tritium Research Laboratory (Sandia – Livermore)

1.0 Introduction

Glovebox confinement of tritium process components and piping protects the workers from radioactive material (especially tritium oxide), provides an inert atmosphere for prevention of flammable gas mixtures and deflagrations, and allows recovery of tritium released from the process into the glovebox when a glovebox stripper system (GBSS) is part of the design. Tritium recovery from the glovebox atmosphere reduces emissions from the facility and the radiological dose to the public. Tritium removal/stripping systems perform a vital part in ensuring that environmental emission compliance and the safety and health of individuals are maintained. Although other technologies and configurations may be available, this review discusses four scenarios for glovebox-stripping systems (GBSSs). These include release with a single or dual stage setup which utilizes either single-pass or recirculation for stripping purposes.

2.0 Background

2.1 Tritium Confinement and Cleanup System Overview

Confinement is defined as "a collection of barriers that can satisfy a specified leak criterion contingent upon operation of its ancillary (active) system. Examples of confinement systems include: a glovebox and its associated cleanup system."[1] Tritium gas process systems are generally enclosed in gloveboxes for confinement. The glovebox allows access to the primary gas processing lines and components for ease of operation and maintenance. The leak rate of a glovebox can generally be certified to be no more than 10^{-3} to 10^{-4} cm^3 He/s. Tritium permeation and diffusion through the elastomeric seals and gloves is reasonably well known, and the tritium released from the glovebox during a primary container leak can be accurately estimated.

If tritium is released into the glovebox, the gases in the glovebox are mixed with and dilute the tritium. The captured tritium is generally removed by circulating the gas through a system that removes tritium down to the part-per-billion level. Typical cleanup systems remove tritium by cracking the molecules on a hot catalyst. The free hydrogen atoms combine in the catalytic reactor with the oxygen present to form water. The tritiated water is then removed from the gas by molecular sieve traps. Depending upon the tritium species, the concentration reduction of these systems can be from one million to one hundred million in a single pass.

2.2 Scope of Glovebox Tritium Stripping Efficiency Review

The GBSS efficiency is assumed to be at least 99%. Verification of this value will useful for the design of related applications. One such application for GBSS is for cleanup of a tritium purification system that is being designed for use in the SHINE Medical Technologies™ production of molybdenum 99. In the SHINE project, a conventional tritium removal/stripper system has been proposed. A conventional tritium cleanup (removal/stripping) system catalytically oxidizes tritium to water which is subsequently trapped on molecular sieves [2-5]. Early calculations utilize a 99% stripping (decontamination or tritium removal) efficiency in regards to permitting and licensing. This literature review evaluates the reported efficiencies of several tritium cleanup systems, and discusses their limitations.

References that collaborate the 99% stripping efficiency are available in publically searchable literature reviews and within the OSTI database. This document details some of the findings of a search for related information. Single pass stripper systems including results from the Idaho National Laboratory, the Mound Laboratory, and the Tritium Research Laboratory of Sandia Laboratory are overviewed. Details of recirculation systems found at the Savannah River Site in applications of conventional stripping and catalytic oxidation are also discussed.

3.0 Single Pass Stripper System

A single-stage single-pass tritium stripper provides the worst case scenario for potential tritium release to the environment in the event of equipment failure. Facilities (historically or currently operated) which describe experimentally based calculations of this type of configuration include the STAR Facility operated by INL, the Mound Laboratory which previously operated as a Department of Energy site, and Sandia Livermore Tritium Research Laboratory. Others are likely, but, reports were not easily found and are therefore not discussed.

3.1 STAR-Facility INL [4]

A single-pass tritium cleanup system was specifically mentioned for the Safety and Tritium Applied Research (STAR) facility operated at the Idaho National Laboratory (INL, formerly INEEL) [4]. In that system, gaseous tritiated effluent from gloveboxes was processed. The operation utilized a configuration shown in Figure 3-1.

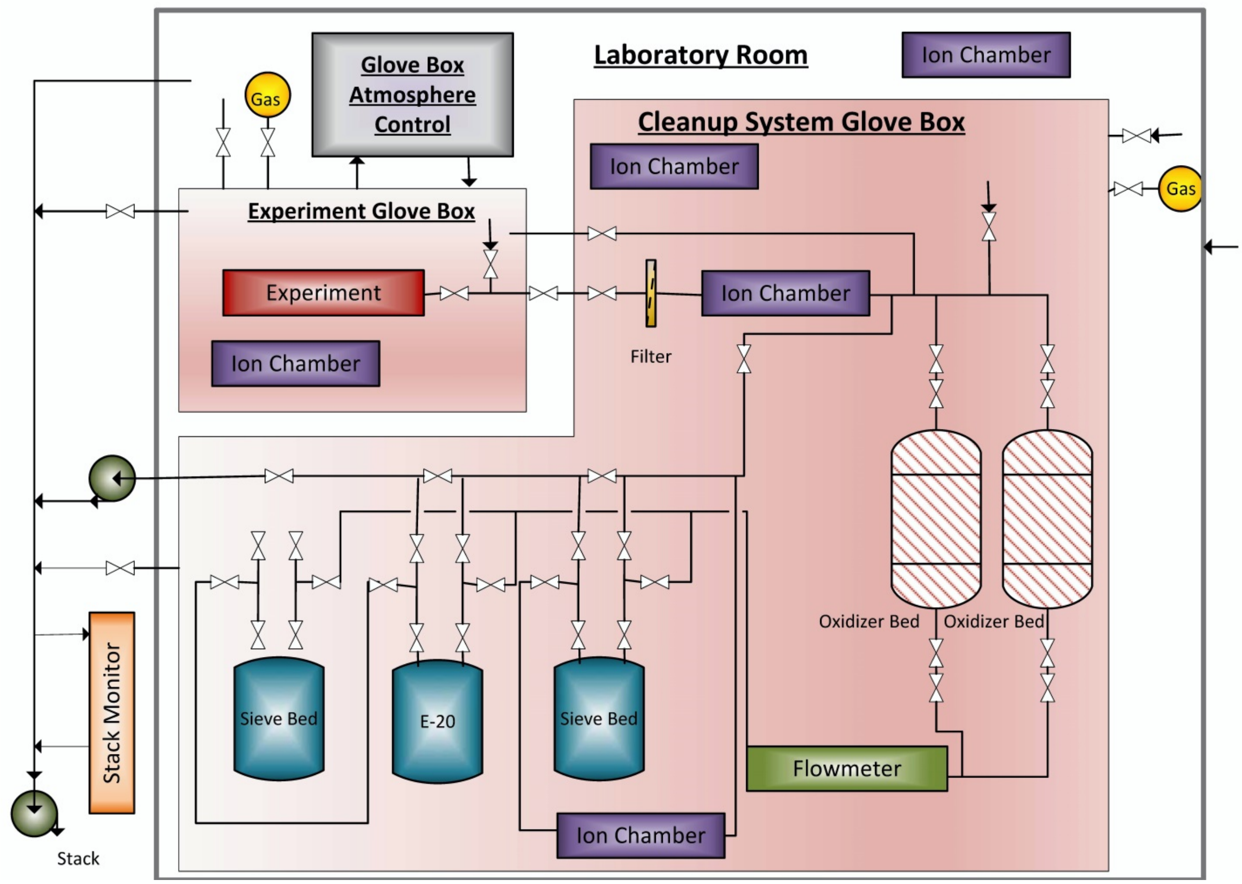

Figure 3-1. Illustration of tritium cleanup system for the STAR facility INL [6].

The tritium cleanup system is described as

two oxidizer beds; two in-line heat exchangers; two molecular sieve bed pairs; two blowers; interconnecting plumbing and isolation valves; diagnostics on inlet and outlet sample loops to measure tritium concentrations, gas flow rates and system pressures; and moisture sensors between each molecular sieve bed pair to measure moisture breakthrough for the first bed.[4].

A palladium catalyst was mentioned and its removal efficiency was found to be related to temperature [4]. An H_2 to H_2O single-pass performance test for one of the oxidizer beds with a pair of molecular sieves at a gas flow rate of 3-7 L/min and an H_2 concentration from 0.8 to 5% at a bed temperature 300°C showed performance efficiency better than 99.9%. A higher flowrate of 10-30 L/min of H_2 concentration from 0.6 to 4%, with oxidizer bed temperature 450°C, showed performance efficiency better than 99.99%. However, at temperatures below 130°C result in capture efficiency far less than 99.9% wherein at 25°C an 11% H_2 removal efficiency was found [4]. This facility did not directly show hot test work (using tritium) however, other sites did describe hot test work. The capture efficiency is still related to a worst case scenario that may be present.

3.2 Mound Laboratory [7]

Many systems discussed in literature tend to have a recirculation system with a single-pass emergency system in place. The emergency containment system of Mound Laboratory was designed to provide the oxidation and adsorption capacity for a single-pass decontamination efficiency of 99.9% in a $2.8x10^4$ L/min air flow containing as high as 1 Ci/m^3 tritium and 0.5 ppm natural hydrogen background [8].

Generally tritium operations underwent a recirculation bed type configuration. In recirculation mode system efficiency was approximately 99.98% using a palladium catalyst at 400°C for a tritium level in air of approximately 0.02 ppm. In recirculation mode a decontamination factor of as high as 3.3 million or 99.99997% was observed during studies [7].

Design of the emergency containment system was undertaken using a series of tests to determine rate kinetics and other parameters. The emergency containment system also operated using a palladium catalyst on inert alumina substrate. They found that the catalyst could give 99.9% conversion in a single-pass at temperatures greater than 177°C with tritium [7].

3.3 Tritium Research Laboratory (TRL) Sandia Livermore [9]

Researchers of the Sandia Livermore, tritium research laboratory decontamination system presented data on containment systems for tritium. This included normal operation in recirculation mode as well as a single-pass concentration reduction system. Research was initiated due to questions regarding possible decontamination efficiency related to nuclear licensing prerequisites at the time. The Mound Laboratory in Ohio (previously discussed in section 3.2) also performed test work in an effort to show the safety of single-pass worst case scenario. Both facilities showed that the palladium catalyst was adequate to meet tritium emissions requirements.

The Gas purification system processed both accidental release as well as slow background tritium concentrations." "The system consists of a central manifold connected to the laboratory gloveboxes, a catalytic reactor, two molecular sieve dryers in series, a blower to circulate the glovebox atmosphere through the system and a control and diagnostics system. Normal operation was in recirculation mode in which the gases of the gloveboxes were pumped from the boxes through the purification system and back to the box until tritium contamination was reduced to acceptable levels [9].

Tritium test of the gas purification system established single-pass concentration reduction factors over a range of inlet tritium concentration from 0.14 Ci/m^3 to 115 Ci/m^3[9].

Both tritiated hydrogen and tritiated methane cases were studied. Tests were able to achieve concentration reduction or decontamination factors greater than 1000 (> 99.9%) per pass with catalysts operated at temperatures in excess of 92°C and 477°C for hydrogen and methane respectively. Hydrogen tests were conducted at an inlet concentration of 100 ppm and a system flow rate of 16.4 std m^3/hr. At 29°C (the lowest temperature achievable the exhaust concentration was below 0.1 ppm. The goal of the test was to demonstrate that reduction factors greater than 1000 were possible [9].

4.0 Recirculation Configuration

4.1 Conventional Stripping

A recirculation stripping system is described for the Replacement Tritium Facility at the Savannah River Site [10-12]. An illustration of this system is found in Figure 4-1. Following a tritium leak to the confinement system, incident data was collected for a recirculation system in which a catalyst bed of palladium deposited on alumina is used before the zeolite bed. Accounting for residual tritium in the system, the concentration readings in the nitrogen decreased with a 97% recovery in the first hour and to 99% recovery within 10 hours [10].

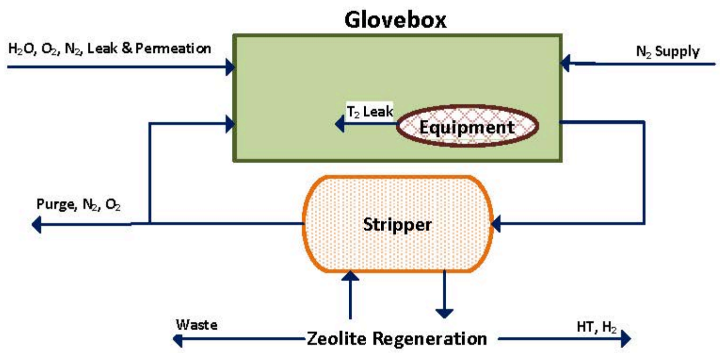

Figure 4-1. Conventional Recirculation System [11]

4.1 Catalytic Exchange

A different configuration from a conventional stripper system configuration to a catalytic exchange stripper was also presented by researchers of the Savannah River Site. An illustration of the differences between the configurations is shown in Figure 4-2.

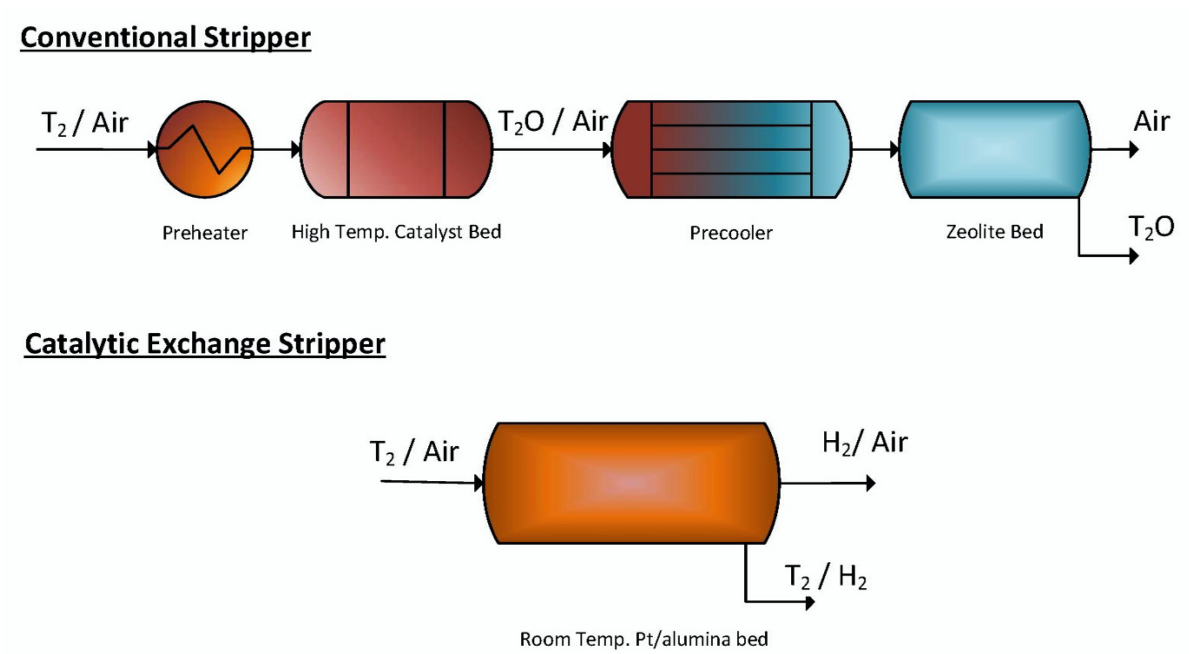

Figure 4-2. Conventional vs catalytic-exchange stripper [13]

The primary difference of the system comes from the fact that a conventional system oxidizes tritium in a catalyst bed and then relies on a molecular sieve to capture the tritium-containing moisture. The catalyst exchange configuration operates at lower temperatures and does not oxidize tritium. Recovery was 99.9% or better using a column packed with alumina pellets containing about 0.3 wt% platinum [13]. The effectiveness of this system are limited to concentrations of 10^4 μCi/m^3 or higher [13]. The dependence of the decontamination factor (DF) on feed concentration (F) in a nitrogen carrier gas was given by:

EQ 1

$$DF = 0.00076F^{0.65},$$

for F > 6.3×10^4 μCi/m^3. Within an air carrier gas stream, the dependence of DF on F is given by

EQ 2

$$DF = 0.0033F^{0.59},$$

for F > 1.6×10^4 μCi/m^3.

Hydrogen removal efficiency is calculated by:

EQ 3

$$Removal\ Efficiency = \left(1 - \frac{1}{DF}\right) * 100\%.$$

5.0 Conclusions

Different tritium decontamination (i.e. cleanup) systems and their related capture efficiencies have been described. Temperature and tritium concentration have been found to be related to capture efficiency. Based on the efficiencies detailed, for both single pass and recirculation stripper systems in this report, the 99% capture efficiency is a reasonable estimate for various configurations of a GBSS with a safety factor included. Tests validated a higher level of decontamination previously during the approval to operate following startup of other U. S. Department of Energy facilities indicated in this document.

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7.0 Acknowledgements

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