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V.P. Gede, Lawrence Livermore Laboratory
P.D. Gildea, Sandia Laboratory Livermore

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TRITIUM PRACTICES PAST AND PRESENT*

V. P. Gede

Lawrence Livermore National Laboratory

P. D. Gildea

Sandia National Laboratories, Livermore

ABSTRACT

Since the discovery of tritium 4 years ago, its production and use have been directed primarily to areas affecting national security. A complete knowledge of tritium technology is required for the world's developing fusion energy program, however, much of the information is not readily available because of classification. Handling techniques first used at Lawrence Livermore National Laboratory made use of glass vacuum systems and relatively crude ion chambers for monitoring airborne activity. The first use of inert atmosphere glove boxes demonstrated that uptake through the skin could be a serious personnel exposure problem. Growing environmental concerns in the early 1970's resulted in the implementation by the Atomic Energy Commission of a new criteria to limit atmospheric tritium releases to levels "as low as practicable." An important result of the new criteria was the development of containment and recovery systems to capture tritium rather than vent it to the atmosphere. The Sandia National Laboratories, Livermore, Tritium Research Laboratory containment and decontamination systems are presented as a typical example of this technology. The application of computers to control systems is expected to provide the greatest potential for change in future tritium handling practices.

A review of the physics literature of the 1930's also establishes the beginnings of thermonuclear theory and experimentation (4). A 1954 paper by W. R. Arnold, et al., established that all fusion reactions between the hydrogen isotopes H, D, and T produce heavier nuclides (5). The reactions were always accompanied by a reduction of the rest mass of the products with the creation of a significant amount of energy (refer to Table I). The activation energies for the hydrogen fusion reactions were generally found to be small.

TABLE I. HYDROGEN FUSION REACTIONS

$H + H \rightarrow D + 0.43 \text{ MeV}$
$H + D \rightarrow {}^3\text{He} + 5.49 \text{ MeV}$
$H + T \rightarrow {}^4\text{He} + 19.8 \text{ MeV}$
$D + D \rightarrow H + T + 4.03 \text{ MeV}$
$D + D \rightarrow {}^3\text{He} + n + 3.27 \text{ MeV}$
$D + D \rightarrow {}^4\text{He} + 28.83 \text{ MeV}$
$D + T \rightarrow {}^4\text{He} + n + 17.57 \text{ MeV}$
$T + T \rightarrow {}^4\text{He} + 2n + 14.32 \text{ MeV}$

Past Practices

PAST TRITIUM PRACTICES

History

In 1934, at Cavendish Laboratory, the work of E. Rutherford, M. L. Oliphant, and P. Harteck produced two mass-three isotopes, ${}^3\text{H}$ and ${}^3\text{He}$, the world's first known synthesis of tritium (1). In 1939, at U. C. Berkeley, L. W. Alvarez and R. Corngood proved that ${}^3\text{He}$ occurred in nature and was stable (2). They later showed that tritium was radioactive (3). The work of these five scientists essentially marked the birth of tritium technology.

Lawrence Livermore National Laboratory (LLNL), in California, began working with tritium late in 1952 when a demand for a source of 14-MeV neutrons ($D + T \rightarrow {}^4\text{He} + n + 17.57 \text{ MeV}$) (5) arose as a consequence of the infant laboratory's entry into thermonuclear weapons research. LLNL's first tritium system was built specifically to tritium-load targets, which were used on a newly completed Cockcroft-Walton accelerator. The standard laboratory-scale glass handling system was comprised of a mechanical vacuum pump for removing air from the system down to a 10^{-5} atmosphere residual pressure, a mercury-filled Toeppler pump for transferring tritium to and from the target loading chamber (7), a mercury man-

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ometer to measure pressure, and a uranium trap to store the tritium. The tritium was produced in the Hanford reactors and supplied to LLNL in 250-ml tubes. Each tube contained 60-120 cc atm of tritium, and was provided with one break-off seal. Good glass-blowing techniques provided the only controlled means of attaching and releasing the tritium to the system. The small system was housed in a wood "piano box" and provided with glass doors and an exhaust blower of approximately 30 cfm capacity.

Tritium control in this small, simple system proved to be very effective. Tritium accountability by PVT techniques was reliable and adequate for management needs. System repair or modification was quick to accomplish and easy to check out. Glass systems were used for well over a decade at LLNL. If given the proper care and maintenance, these glass systems could be used to this day.

Steel dry-boxes were put into service at LLNL in 1952. These boxes were designed to take a total vacuum and a rapid pump-down. They were generally operated at 1 atm argon pressure. No provision was made for the removal of water vapor, oxygen, and nitrogen which permeated through the gloves and elastomeric seals to the box interior. Frequent box pump-downs were required to maintain even a 99 percent argon atmosphere.

The first LLNL tritium box-line was put into service in mid-1953. This box-line consisted of three boxes in series connected to a double-ended air lock, which in turn was tied to a special machining box. The box-line, made of stainless steel, was equipped with numerous windows and viewing ports of one-inch-thick tempered glass. There were also many glove ports provided with heavy, 0.030-in. thick rubber gloves.

This facility was used for handling solid, reactive tritium compounds. Personnel exposures became a concern immediately after the introduction of tritium into the facility. Frequent box-line evacuations provided some immediate relief, but had a short-term overall effect.

Operations requiring grinding and compounding in the box caused severe dusting. This dusting put a film of reactive tritiated material on the inner box-line surfaces. This material readily reacted with oxygen and moisture diffusing into the box-line through the rubber gloves and gaskets, and brought the tritium level of the box-line atmosphere up to undesirable concentrations in a few hours.

The box-line's tritium concentration was determined by diverting the contaminated air-lock atmosphere through the air lock vacuum pump to a room air monitor. Argon was used to purge the tritium activity from the pump exhaust through the ioniza-

tion chamber to the facility exhaust. By assuming a constant argon purge-flow time, and knowing the ratio of box-line volume to air-lock volume and the ionization chamber reading, a calculation of box-line atmosphere tritium content could be made.

The tritium monitors used during those times were crude but effective ionization chamber instruments. These instruments gave a reliable reading of room air tritium activity. The main complaint in those days was that the monitors were excellent in showing an increase in activity (large or small), but after reading a large increase the monitor took a long time (many minutes) to show any diminution in activity. This fault occurs even with today's more sophisticated tritium monitors. Urinalyses were routinely run on all tritium laboratory personnel. Tritium body burdens of 30 μ Ci/l were accepted maximums.

The lessons learned with the early tritium systems were, at least in hindsight, predictable. As a result of radiation damage, the vacuum grease, which is critical to effective operation of the glass vacuum system, turned to a hard rubber-like substance. This froze the stop cocks and taper seals, and elastomer materials, such as neoprene o-rings and gaskets, became inflexible and lost sealing effectiveness. Mass spectrometric analyses of a given tritium sample showed that the protium content increased continuously. On unexposed surfaces this isotopic exchange reaction readily occurs. Sample vessels with specially conditioned surfaces are used to help eliminate this problem. Tritium does an extremely effective job of equilibrating with the accessible protium/deuterium in a tritium system.

The use of commonly accepted materials for construction of tritium systems, copper, aluminum, and austenitic stainless steels, is strongly recommended. Stainless steel systems should be designed for low working stresses to avoid brittle failure. Reaction products generated by exposure of elastomeric materials (such as teflon and polyethylene to tritium) can produce corrosion of many materials, and intergranular stress corrosion cracking in stainless steels. This failure mechanism was verified by tests of highly stressed Vin Var stainless steel pressure vessels at LLNL in 1978 (8).

PRESENT TRITIUM PRACTICES

For purposes of this paper, "Present Tritium Practices," refers to handling technology that has originated primarily within the past eight to ten years and that is being applied to facilities presently under construction. Until this period the majority of tritium operations in the U.S. were conducted in high-velocity air hoods, and any tritium released was vented to the atmosphere through the facility stack. Approximately 10 years ago, environmental

concerns prompted the Atomic Energy Commission (AEC) to adopt the philosophy of limiting atmospheric releases to levels "as low as practicable" (ALAP). Pressure was applied to U.S. tritium facility operators to institute programs to reduce tritium releases. It should be emphasized here that the principal concern 10 years ago was for environmental protection. It had been demonstrated many years prior to this that high-velocity air hood operations provided adequate personnel exposure protection.

The AEC program prompted the operators of tritium facilities, and any contractors seeking authorization for new tritium facilities, to look at methods for containing tritium releases and recovering the tritium rather than venting it to the atmosphere. Although a small effluent cleanup system had been in operation at General Electric Co. Neutronic Devices Department since 1960 (9) the first effluent cleanup system of significant capacity (i.e., 102 m³/hr) was put into operation at the Mound Facility in 1972(10) as a response to the AEC ALAP policy. Table II summarizes the tritium recovery systems presently in operation and under construction. In my opinion, containment and recovery systems have been the principal new tritium handling technology to emerge from the past decade. This paper will review the design and operation of containment and removal systems that have been put into operation or designed in the past five years, and will only briefly consider transfer and storage technologies in the discussion of future developments. This time frame has been chosen to emphasize the operation and design of relatively large systems providing some type of centralized facility protection, not just isolated glove box-line protection. The Sandia National Laboratories, Livermore (SNLL), Mound Facility, Los Alamos Scientific Laboratory (LASL),

and Princeton Plasma Physics Laboratory (PPPL) have put such systems into operation or have them under construction.

It should be noted that the technology for these systems is not new. Although none of these systems are identical, they all use the same commercially available building blocks, i.e., blowers, catalyst beds or recombiners, and dryers. Flow rates range from fractions of m³/hr to thousands of m³/hr. Catalysts, adsorbents, and operating temperatures vary with the system requirements. Rather than attempt to describe the operation of all of these systems, I shall focus on the SNLL Tritium Research Laboratory (TRL) containment and decontamination systems as they are representative of the philosophy and hardware in use, and I am much more familiar with them.

SNLL CONTAINMENT AND DECONTAMINATION SYSTEMS

About ten years ago Sandia National Laboratories (SNL) began seeking AEC authorization for construction of a tritium facility at the Livermore site. To meet ALAP criteria, SNL proposed a new approach using secondary containment in conjunction with central tritium removal systems. Construction of the TRL (11), designed for a wide range of experiments using multi-gram amounts of tritium, was authorized in 1973, and entered its start-up phase on October 1, 1977. In addition to the usual pressure-gradient-controlled once-through ventilation system, the TRL provides for both personnel safety and environmental protection by employing a glove box secondary containment system connected to two central decontamination systems, the gas purification system (GPS) and the vacuum effluent recovery system (VERS). Since the VERS uses the same tritium removal technology as the GPS, only the GPS system will be discussed in this paper.

Containment System Design and Operation

All experiments containing significant amounts of tritium are secondarily contained in sealed, stainless steel glove boxes of welded construction. A typical glove box is illustrated in Figure 1. Each box has glove ports, viewing windows, an air lock pass-through, and removable panels on each end for the installation of large items. Regular utility and emergency electrical power are provided, as well as feed-through provisions for instrumentation, inert gas pressure connections, and a cooling system to remove the heat generated by experiments. Tritium concentration and humidity control can be maintained by processing the glove box atmosphere through the GPS. Normally, the glove boxes are operated with dry nitrogen maintained by the box pressure control system at -0.25 to -1.0 kPa, with respect to the room. The GPS assumes the pressure

TABLE II. CLEANUP SYSTEM SUMMARY

System Location	Year Put in Operation	Capacity (m ³ /hr)	Function
General Electric Co. Neutronic Services Department	1960	6-12	Effluent cleanup (9)
Livermore Laboratory Tritium Facility	1967	95-80	Dry box cleanup
Mound Facility	1972	102-66	Effluent cleanup (10)
Los Alamos Scientific Laboratory	1972	6-1	Dry box cleanup (12)
		25-1	Effluent cleanup (13)
Mound Facility Tritium Effluent Cleanup Laboratory	1977	25-1	Air atmosphere (16) glove box cleanup
		158-5	Nitrogen atmosphere (16) glove box cleanup
		1760-67	Room air cleanup (16)
Sandia Laboratories, Livermore Tritium Research Laboratory	1977	17-60 340-60	Effluent cleanup (12, 13) Glove box cleanup (12, 13)
Los Alamos Scientific Laboratory Tritium Systems Test Assembly	1978	90-00 2340-00	Effluent cleanup (17) Room air cleanup (18)
Princeton Plasma Physics Laboratory Tokamak Fusion Test Reactor	1981	17-00 1500-00	Glove box cleanup (19) Tritium cleanup (19)
Under construction			Room air cleanup (19)
References			

control function for the glove box during the clean-up cycle. The box can also be operated with an argon or air atmosphere, if desired.

Fig. 1. Sealed glove box.

Decontamination System Design and Operation

The GPS (12), illustrated in Figure 2, removes tritium from the glove box atmosphere in the event of an accidental release, or a slow buildup of background concentration. The GPS consists of a central manifold connected to each of the laboratory glove boxes, a catalytic reactor to oxidize the tritium, two molecular sieve dryers in series to collect the tritiated water, a blower to circulate the glove box atmosphere through the system, and a control and diagnostics system to provide both automatic and manual control and to assess operational status. Redundant prime function components, i.e., blowers, heaters, and catalyst and dryer beds are provided for reliability. Moisture swamping is used for the downstream dryers. A dryer regeneration system provides removal of water from a loaded dryer while the GPS is operating with the use of the other two dryers. Entry and exit nozzles allow safe removal and replacement of the catalyst and sieve material while the system remains operational by use of a redundant flow path. Major system performance criteria are summarized in Table III.

Fig. 2. GPS Schematic

TABLE III. GPS PERFORMANCE CRITERIA

Processing Capacity	340 m ³ hr ⁻¹
Catalytic Reactor	
Operating Temp.	783 K
Catalyst	Engelhard Minerals and Chemicals No. A-16648
Tritium Conc.	1000 per pass for conc.
Reduction Factor*	from 20,000 to 1 ppm
Molecular Sieve Dryer	
Capacity	37 moles of water while maintaining < 1 ppm water at exit
Molecular Sieve	Type 4A
Startup Operating Time	30 s

*Ratio of inlet to outlet tritium concentrations.

The GPS can be operated either in a recirculation mode or in a stacking mode. In the recirculation mode, the normal method of operation, the glove box gases are pumped from the box, through the GPS, and back to the box until tritium contamination is reduced to an acceptable level. The stacking mode is similar to the recirculation mode with the exception that the glove box atmosphere is not recirculated, but stacked after passing through the GPS. This is accomplished by drawing clean box-atmosphere gas into the manifold ahead of the glove box and venting the GPS effluent to the ventilation exhaust. This mode of operation may be used to backfill the glove box with dry nitrogen or argon when an inert atmosphere is required. It can also be used to drive the glove box concentration to low levels when it is necessary to clean up a box for reentry.

The GPS is controlled by a stored-program controller utilizing an Intel 8080 microprocessor with EPROM memory. The controller is interfaced to the DEC 1140 facility computer, and has been programmed to initiate glove box cleanup on command from the DEC computer. The controller selects components as a function of the operational status, positions the automatic valves to establish an open path, powers up the selected components to establish flow, adjusts the manifold pressure, and signals the 1140 when it is ready for operation. The DEC 1140 then opens the valves to the selected glove box to initiate tritium removal. The GPS is also equipped with a manual control panel that may be used to operate the GPS in the event of computer failure.

Decontamination System Performance

The performance of the GPS was evaluated through a series of tests (13). The first tests were run with hydrogen and methane to examine the effect of temperature upon the catalyst conversion efficiency. The results, displayed in Figure 3, determined that a catalyst operating temperature of 780 K was required to achieve a concentration reduction factor of 1000 for methane. Final performance tests were run with tritium to determine the catalyst conversion efficiency. The results presented in Table IV show that concentration reduction factors of 10⁴ and greater can be achieved at inlet concentrations of less than one ppm tritium. Finally, a glove box cleanup test was run with methane at an initial concentration of 500 ppm to determine the rate a contaminant could be removed from the glove box atmosphere. The results, presented in Figure 4, show that the GPS can achieve a cleanup rate approaching exponential dilution.

Fig. 3. Effect of GPS Catalyst Temperature on Concentration Reduction Factor

TABLE IV. GPS TRITIUM TEST SUMMARY

Test Designation	Sample Composition	Inlet Tritium Concentration $\mu\text{Ci m}^{-3}$	Exhaust Tritium Concentration $\mu\text{Ci m}^{-3}$	Concentration Reduction Factor
GPS T-1	T ₂ in nitrogen	0.14	2.0	7.0×10^4
GPS T-2	T ₂ in nitrogen	1.3	15.0	8.7×10^4
GPS T-3	T ₂ & 2.01 H ₂ in nitrogen	9.4	1020.0	9.2×10^3
GPS T-4	T ₂ in nitrogen	115.0	250.0	4.6×10^5

Fig. 4. Glove Box Cleanup Test Results

DESIGN FEATURES OF OTHER SYSTEMS

There are a total of ten cleanup systems, (refer to Table II), other than those in the TRL, that have been put into operation or designed within the past five years. Some of the notable design features of these systems that differ from the TRL system are summarized in Table V. The liquid nitrogen-cooled molecular sieve bed is probably applicable only to helium atmosphere systems; however, the other features may be used in any system where they provide an operational advantage.

TABLE V. NOTABLE DESIGN FEATURES OF OTHER SYSTEMS

System	Design Feature	Purpose
Mound Facility He Atmosphere Glove Box Cleanup	Liquid nitrogen-cooled molecular sieve adsorbent	Remove air contaminants and tritium from He glove box atmosphere
Mound Facility Room Air Cleanup	Prenaturated adsorption/ exchange column using Alcoa alumina H181 adsorbent	Remove oxidized tritium
Los Alamos Scientific Lab Tritium Systems Test Assembly Room Air Cleanup	Refrigerated condenser for water removal prior to molecular sieve bed	Extend dryer life
Princeton Plasma Physics Laboratory All Systems	Cooler between molecular sieve stages	Increase second dryer efficiency

FUTURE DEVELOPMENTS

The greatest potential for change in tritium handling technology lies in the application of compu-

ters, particularly microprocessors, to control systems. I feel this applies as equally to transfer and storage systems where the fundamental hardware and handling techniques have not markedly changed in 20 years, as to the containment and cleanup systems that are emerging as a result of environmental concerns.

Most past tritium releases have resulted from material failures or human error. Containment and cleanup systems remove the majority of the risk of material failure. As a result of the capability to define predetermined flow paths and build in safety-interlocks, computer operated-or-aided control systems will virtually eliminate tritium releases due to operator error.

The SNLL TRL, PPPL, and LASL facilities all contain to some extent computer-based control systems. However, LASL is the only facility whose planning now calls for completely computer-controlled operation and generation of safety-related diagnostics.

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