2/
6/13/78

SAND77-8766
Unlimited Release

8265, PUBLICATIONS AND PUBLIC INFORMATION
DIV. FOR TIC (2)

CONF-780508--16

Results of Tritium Tests Performed on Sandia Laboratories Decontamination System

(Presented at The Technology of Controlled Nuclear Fusion Conference held May 9-11, 1978 in Santa Fe, NM and to be published in the proceedings)

MASTER

P. D. Gildea, W. R. Wall, V. P. Gede

Sandia Laboratories

SF 2900 Q(7-73)

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

SAND [illegible]
Unlimited Release
Printed May 1978

RESULTS OF [illegible] ON SANDIA LABORATORIES DECONTAMINATION SYSTEM

[illegible] and

SANDIA LABORATORIES
LIVERMORE, CALIFORNIA 94550

V. P. [illegible]

LAWRENCE LIVERMORE LABORATORY
LIVERMORE, CALIFORNIA 94550

The Tritium Research Laboratory (TRL), a facility for performing experiments using gram amounts of tritium, became operational on October 1, 1977. As secondary containment, the TRL employs sealed glove boxes connected on demand to two central decontamination systems, the Gas Purification System and the Vacuum Effluent Recovery System. Performance tests on these systems show that tritium removal systems can achieve concentration reduction factors (ratio of inlet to exhaust concentrations) much in excess of 1000 per pass at inlet concentrations of 1 part per million* or less for both tritium and tritiated methane.

INTRODUCTION

The data presented in this paper are the result of performance tests completed on the two central decontamination systems incorporated in the Sandia, Livermore, Tritium Research Laboratory[1] (TRL). The laboratory (Figure 1), designed as a research facility for a wide range of experiments using gram amounts of tritium, became operational on October 1, 1977.

The Tritium Research Laboratory provides for both personnel safety and environmental protection by employing a secondary containment system connected to two central decontamination systems, the Gas Purification System (GPS) and the Vacuum Effluent Recovery System (VERS).

CONTAINMENT

All experiments are secondarily contained in sealed stainless steel glove boxes of welded construction (Figure 2). Each box is equipped with glove ports, viewing ports, and an air lock pass-through; and on each end there are removable panels for the installation of large items. Both regular utility and emergency electrical power are provided within the box. Also, there are feedthrough provisions for instrumentation and for inert gas pressure connections, and a cooling system

---

* 1 part per million = 2.6 Ci m^{-3} for T_2 and CH_2T_2.

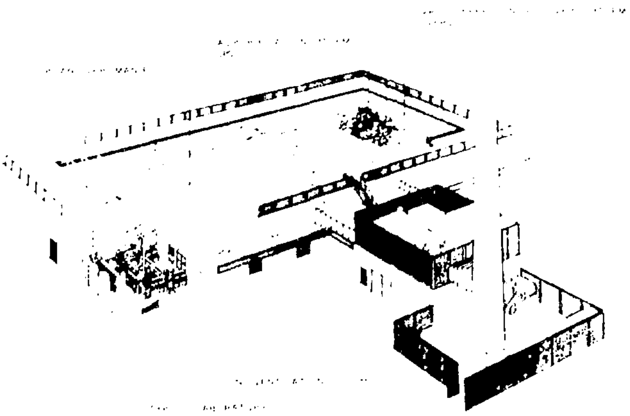

Figure 1. The TRL Building at Livermore, Showing the Arrangement of Some of the Major Systems and a Typical Experiment Station

to remove the heat generated by experiments. Both tritium concentration and humidity control are maintained by connecting the boxes to the decontamination system. Normally, the glove boxes are operated with dry nitrogen maintained by the box preseure control system at a pressure of -0.25 to -1.0 kPa with respect to the room; however, the box can be operated with an argon or air atmosphere if desired.

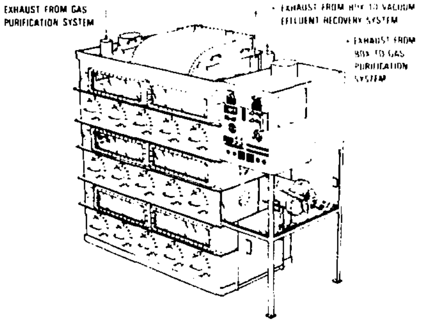

Figure 2. Sealed Glove Box

DECONTAMINATION

Decontamination is accomplished by two centralized systems.[1] The first of these, the GPS, removes tritium and tritiated hydrocarbons from gases exhausted from the laboratory vacuum systems before the decontaminated residue gases are vented to the stack. Both systems were designed* to be capable of reducing tritium concentrations to the low parts-per-billion level. All of the tritium removed by the decontamination systems is contained, either for recovery or for disposal as solid waste on type 4A molecular sieves.

Gas Purification System

The GPS (Figure 3), a 340 m^3 hr^{-1} flow-rate system, which protects against either an accidental release or the slow buildup of background tritium concentrations, consists of a central manifold connected to each of the laboratory glove boxes, a catalytic reactor to oxidize the tritium, two

---

* Original systems were designed by Engelhard Industries Systems Department, Union, NJ

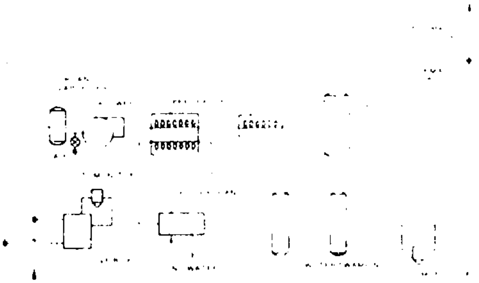

Figure 3. Gas Purification System Schematic

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 remote and local control and to assess operational status. In the recirculation mode, the normal method of GPS 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.

Vacuum Effluent Recovery System

The VERS (Figure 4), a 17 $m^3 hr^{-1}$ flow-rate system, which decontaminates exhaust gases from the laboratory vacuum systems before venting to the stack, is similar to the GPS and uses the same precious metal catalyst.* The system consists primarily of a laboratory vacuum manifold and two holding tanks to collect the contaminated waste gases, a catalytic reactor to oxidize the tritium, two molecular sieve dryers connected in series to collect the tritiated water, the necessary pumps to evacuate the laboratory manifold and transfer the waste gas through the system, and a control and diagnostics section to provide both automatic and manual operation and to assess operational status.

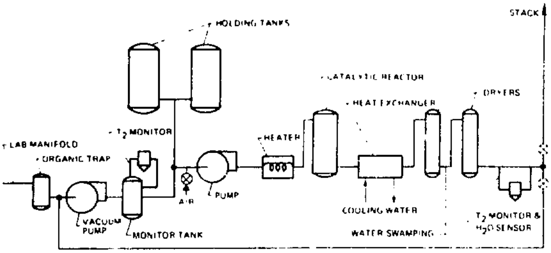

Figure 4. Vacuum Effluent Recovery System Schematic

---

* Engelhard Catalyst No. A 16648

Both the VERS and GPS catalytic reactors are maintained at an elevated temperature (783K) in standby condition to ensure readiness to begin processing upon receipt of a start signal.

DECONTAMINATION SYSTEM TESTING

The decontamination systems in the TEL were given two series of performance tests. The first series was with hydrogen and methane to provide confidence that the catalytic reactors in both systems were operating properly before tritium was introduced. The tests were also to determine the appropriate catalyst operating temperature.

The second test series was with tritium and tritiated methane to determine that the systems were capable of achieving the design requirement of a single-pass concentration reduction factor (ratio of inlet to exhaust concentration) of 1000 per pass for inlet concentrations of 1 part per million. Sensitivity limitations of gas chromatography required that this confirmation be made with tritium as the test gas. Tests were run with methane and tritiated methane because tritiated hydrocarbons are expected to be present in both systems,[2] particularly in the VERS.

GPS Hydrogen and Methane Tests

Many runs were made to assess the system operation before the parameters shown in Table I were selected for tests measuring temperature effects upon catalyst performance. Inlet concentrations were chosen to provide a reasonable range of detection for the gas chromatograph.

TABLE I

GPS HYDROGEN AND METHANE TEST PARAMETERS

Test Gas	Flow Rate std m^3 hr^{-1}	Inlet Concentration ppm	Catalyst Temperature K
Hydrogen	340	120	316-743
Methane	261	700	316-810

The test gas, together with oxygen, was injected into the manifold upstream of the system blowers. The oxygen, which was added to permit combustion, was injected in an amount exceeding stoichiometric requirements. (A simplified flow diagram of the test configuration is shown in Figure 5.) Inlet and exhaust concentrations were measured with a gas chromatograph both upstream and downstream of the catalytic reactors at the approximate locations of the tritium monitor sampling lines.

The effect of temperature upon the concentration reduction factor is displayed in Figure 6

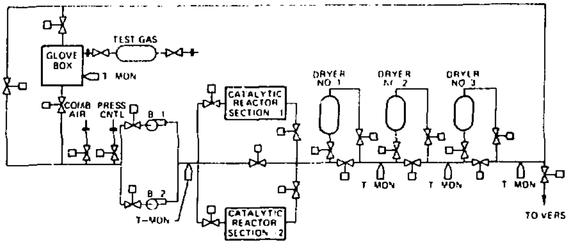

Figure 5. GPS Tritium Test Configuration

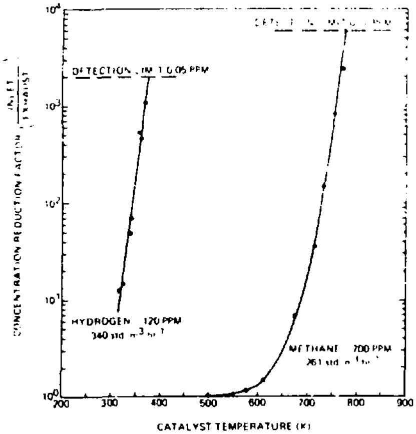

Figure 6. Effect of Catalyst Temperature on Concentration Reduction Factor

for both hydrogen and methane. Temperatures below 316K were not achievable because of the heat of compression generated by the circulation blowers.

GPS Tritium Tests

A series of four tritium tests has been completed to date. The test parameters are summarized in Table II.

TABLE II
TRITIUM TEST PARAMETERS

Test Designation	Sample Composition	Total Tritium Curies	Flow Path
GPS T-1	Tritium in Nitrogen	1.0	Blower 1 Catalyst 1 Dryers 1 and 2
GPS T-2	Tritium in Nitrogen	10	Blower 2 Catalyst 2 Dryers 1 and 3
GPS T-3	Tritium and 2% Hydrogen in Nitrogen	75	Blower 2 Catalyst 2 Dryers 2 and 3
GPS T-4	Tritium in Nitrogen	750	Blower 2 Catalyst 2 Dryers 2 and 3

All four tests were run at a catalyst temperature of 783K and a flow rate of 340 std m^3 hr^{-1}. These tests were run without adding water ahead of the second dryer. Hydrogen was added to some test runs to simulate processing tritium at higher concentrations.

In all cases, the test gas was injected into the glove box (Figure 5) by flushing the test gas cylinder with nitrogen; and in the case of GPS T-3, hydrogen was injected into the box to achieve the 2 percent concentration after the tritium injection had been completed. The glove box cooling system blower was running during both injection and testing to ensure complete mixing of the gases in the glove box. Glove box cleanup was terminated when the tritium concentration reached approximately 10 mCi m^{-3} or less. Tritium concentrations were measured in the glove box and at five locations in the GPS. The least counts of the inlet and exhaust tritium monitors were 1 mCi m^{-3} and 1 μCi m^{-3}, respectively. Test results are summarized in Table III.

TABLE III
GPS TRITIUM TEST RESULTS

Test Designation	Maximum Inlet Tritium Concentration Ci m^{-3}	Maximum Exhaust Tritium Concentration μCi m^{-3}	Concentration Reduction Factor
GPS T-1	0.14	2.0	7.0×10^4
GPS T-2	1.3	115	8.7×10^4
GPS T-3	9.4	1020	9.2×10^3
GPS T-4	115	250	4.6×10^5

Concentration reduction factors were calculated by dividing the maximum inlet tritium concentration by the maximum exhaust tritium concentration. Although the reduction factor measured in GPS T-3 is much larger than 1000, it is smaller than expected for the inlet concentration used. VERS test T-6 was also run with 2 percent hydrogen, but the

VERS data are not directly comparable since the catalyst residence time for the VERS is approximately 2.5 times that for the GPS. However, had the VERS T-6 concentration reduction factor been similarly lowered by an order of magnitude, the exhaust tritium concentration would have been about 10 μCi m^{-3}, a detectable concentration. A number of hypotheses have been proposed to explain the anomaly, and a test program is planned to determine the cause of the lower value. An uncertainty of approximately 30 percent should be applied to the GPS concentration reduction factors because of inaccuracies in both tritium measurements and data acquisition methods.

VERS Hydrogen and Methane Tests

Since the GPS and the VERS use the same catalyst material and are operated at the same temperature, it was not considered necessary to repeat all of the GPS testing. It was, however, important to ensure personnel safety by demonstrating that the system would perform as expected before tritium was put into it. To achieve this aim, concentration reduction factors were measured as a function of catalyst temperature for both hydrogen and methane. The test gases, nitrogen and combustion air, were injected upstream of the catalytic reactor, and inlet and exhaust concentrations were measured with a gas chromatograph upstream of the catalytic reactor and downstream of the heat exchanger, respectively (Figure 7). Again, oxygen was injected in excess of stoichiometric requirements.

The hydrogen test was performed at an inlet concentration of 100 parts per million and a system flow rate of 16.4 std m^3 hr^{-1}. At 302K, the lowest temperature achievable, the exhaust concentration was below 0.1 part per million, which is the detection limit of the chromatograph. The methane test was performed at an inlet concentration of 2000 parts per million and a system flow rate of 13.3 std m^3 hr^{-1}. The results were essentially the same as those for the GPS methane test (Figure 6).

VERS Tritium Testing

A series of seven tests was completed on the VERS. Four tests were run with tritium, the remainder with tritiated methane. The test conditions are summarized in Table IV.

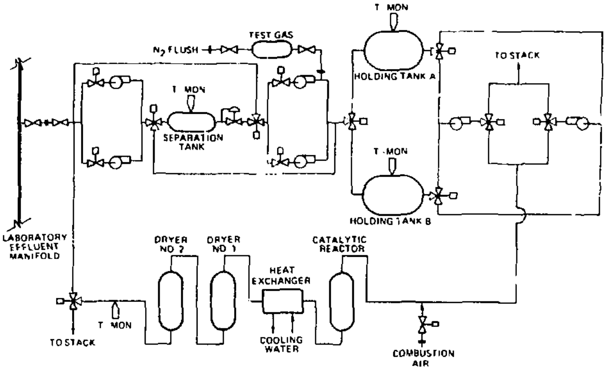

Figure 7. VERS Tritium Test Configuration

TABLE IV

VERS TRITIUM AND TRITIATED METHANE TEST PARAMETERS

Test Designation	Sample Composition	Total Tritium (Ci)
VERS T-1	Tritium in Nitrogen	1.0
VERS T-2	Tritium in Nitrogen	1.0
VERS CH_3T-1	Tritiated Methane in Nitrogen	1.0
VERS CH_3T-2	Tritiated Methane in Nitrogen	10
VERS CH_3T-4	Tritiated Methane and 0.5% Hydrogen in Nitrogen	40
VERS T-5	Tritium and 0.5% Hydrogen in Nitrogen	75
VERS T-6	Tritium and 2% Hydrogen in Nitrogen	750

VERS T-1 and T-2 were identical tests, except that the alternate holding tank and transfer pumps were used. The seven tests were all performed at a catalyst temperature of 783K and a flow rate of 17.0 std m^3 hr^{-1}. As in the case of the GPS tests, these were run without adding water ahead of the second dryer. Also, hydrogen was added to some test runs to simulate processing tritium at higher concentrations.

The test gas was injected downstream of the transfer pump into one of the 5.7 m^3 holding tanks which had been evacuated to 13 kPa (Figure 7). Nitrogen was used to flush the test gas out of the sample cylinder and into the holding tank, and the procedure was continued until the holding tank pressure reached approximately 86 kPa. After the tritium concentration in the tank had stabilized, the holding tank contents were processed through the recovery system and circulated into the alternate holding tank, where the concentration was measured to verify tritium removal before the effluent was stacked. Tritium concentrations were measured in each of the holding tanks and at the outlet of the second dryer. The holding tank tritium monitors and the exhaust tritium monitor had least counts of 10 $\mu Ci\ m^{-3}$ and 1.0 $\mu Ci\ m^{-3}$, respectively. The results are summarized in Table V.

TABLE V

VERS TRITIUM AND TRITIATED METHANE

[illegible]

Test Designation	Inlet Tritium Concentration ($Ci\ m^{-3}$)	Exhaust Tritium Concentration* ($\mu Ci\ m^{-3}$)	Concentration Reduction Factor
VERS T-1	0.18	<1.0	>1.8 x 10^5
VERS T-2	0.18	<1.0	>1.8 x 10^5
VERS CH_3T-1	0.18	<1.0	>1.8 x 10^5
VERS CH_3T-2	1.8	<1.0	>1.8 x 10^6
VERS CH_3T-4	7.4	<1.0	>7.4 x 10^6
VERS T-5	13.2	<1.0	>1.3 x 10^7
VERS T-6	132	<1	>1.3 x 10^8

As in the case of the GPS, the concentration reduction factors were calculated by dividing the maximum inlet tritium concentration by the maximum exhaust tritium concentration. However, during the VERS test, the inlet tritium concentration remained constant. An uncertainty of approximately 50 percent should be applied to the VERS concentration reduction factors because of inaccuracies in tritium measurements at 1 $\mu Ci\ m^{-3}$.

SUMMARY

Tests with hydrogen, methane, tritium, and tritiated methane have been performed on the Sandia Tritium Research Laboratory decontamination systems to determine their ability to meet design specifications.

1. The hydrogen and methane tests show that to achieve concentration reduction factors greater than 1000 per pass, the catalyst must be operated at temperatures in excess of 365K and 750K for hydrogen and methane, respectively.

2. Tritium and tritiated methane tests on the Vacuum Effluent Recovery System have established lower bounds on single-pass concentration reduction factors over a range of inlet tritium concentrations from 0.18 Ci m^{-3} to 132 Ci m^{-3}.
3. Tritium tests of the Gas Purification System have established single-pass concentration reduction factors over a range of inlet tritium concentrations from 0.14 Ci m^{-3} to 115 Ci m^{-3}.
4. An apparently anomalous data point has been generated in GPS T-3, wherein hydrogen was added to achieve a 2 percent mixture to simulate higher tritium concentrations. Additional tests are planned to assess the validity of the data point, even though the reduction factor was considerably greater than 1000.

The test program to date has demonstrated that both the Gas Purification System and Vacuum Effluent Recovery System perform 10-1000 times better than required by specifications and that tritium removal systems can be designed to achieve concentration reduction factors much in excess of 1000 per pass at inlet concentrations of 1 part per million or less for tritium and for tritiated methane.

REFERENCES

1. P. D. Gildea, et al, A Tritium Systems Test Facility Volume II: Appendixes, Sandia Laboratories, Livermore, SAND 76-8053, October 1976, pp. 57-59.
2. J. C. Bixel and C. J. Kershner, "A Study of Catalytic Oxidation and Oxide Adsorption for the Removal of Tritium From Air," AEC Pollution Control Conference, April 16-19, 1974.