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**Tritium Waste Control:
April-September 1977**

January 9, 1978

Monsanto

MOUND FACILITY

Miamisburg, Ohio

operated by

MONSANTO RESEARCH CORPORATION

a subsidiary of Monsanto Company

for the

U. S. DEPARTMENT OF ENERGY

Contract No. EY-76-C-04-0053

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Foreword

The format of this document has been changed to reflect changes in funding. The work in the first three reports are funded by 189C AL0219A. The remaining sections are work related to Tritium Waste Control but not directly funded by AL0219A.

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A pilot scale system was used in an initial experiment to investigate the combined-electrolysis-catalytic-exchange process (CECE) for the detritiation of water. Data taken during the experiment indicate the process does indeed strip tritium from gaseous hydrogen at the top and concentrate it in water at the bottom of the catalyst-filled column. Several deficiencies were found in the system as originally assembled. Modifications are presently under way to correct these deficiencies.

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A high activity tritiated water electrolysis system was designed and built using a General Electric Company solid polymer electrolyte (SPE) cell. The system was successfully operated at currents up to 50 A using deionized tap water. It was found that large amounts of water (60-70 cm³ in 40 min at 40 A) are transferred across the SPE to the hydrogen side of the cell. This will necessitate frequent system shutdowns to drain the hydrogen reservoir.

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Triplicate samples of cement, cement-plaster (1:1 ratio by weight), and cement-plaster (1:1 ratio by volume) were injected with 386 Ci of tritium. All samples were cured for five days and impregnated with catalyzed styrene monomer. After polymerization the samples were put into a test program to measure the tritium release. Preliminary results indicate Type III Portland cement retains the tritium slightly better than the cement-plaster mixtures. After six weeks about 0.4 Ci of tritium is released which is a fractional release of 1.0×10^{-3} from the test package.

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The tritium release study of actual burial packages is continuing. The fractional release is 1×10^{-5} on a 4-yr old package, only 4×10^{-7} on 3-yr old packages, and 1×10^{-9} on a 1-yr old package.

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Pressure increase and gas composition determinations were repeated for octane (activity = 1000 Ci/liter) with and without fixation using argon as the initial overgas. Pressure buildup measurements for octane without fixation were repeated a third time using hydrogen gas. The rate of pressure increase did not change significantly from previously

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Four elemental tritium samples were released into a laboratory to determine the efficiency of the Emergency Containment System to decontaminate the laboratory atmosphere. The initial three releases showed that the dampers in the ventilation system were not adequate to prevent tritium from leaking out of the laboratory and therefore decontamination factors less than 10 resulted. The ventilation system was modified during the fourth experiment to minimize leakage. This experiment resulted in a decontamination factor of 1800 which is comparable to the design criteria.

The Glovebox Atmosphere Detritiation System efficiency to remove tritium was determined by releasing two tritium samples into a glovebox. The calculated efficiency of the system was determined to be 0.56 and 0.77 for the two experiments as compared to 0.99998 which was the design goal.

Catalytic exchange detritiation studies

R. E. Ellis, T. K. Mills, and M. L. Rogers

Background

In the operation of a light water reactor, all radioisotopic contaminants except tritium can be removed from the effluent stream by ordinary chemical and physical methods. Tritium removal in this case requires special considerations. Isotopic separation processes, therefore, need to be developed for removing tritium from contaminated water streams.

The objectives of this study are to:

1. Determine technical and economic factors of HT/H₂O exchange as a process for treating tritium contaminated water;
2. Determine suitability of hydrophobic exchange catalysts for use in water detritiation systems; and
3. Through pilot scale testing, establish design criteria for detritiation systems capable of meeting the tritium control needs of DOE site operations and the U. S. power reactor industry.

Limited operation of the Englehard experimental system indicated that the principles involved in the catalytic exchange process are valid. Use of the system has been discontinued, however, because the experimental error incurred was too large for further operation to be of value.

A preliminary economic evaluation was made of the HT/HTO catalytic exchange detritiation stripping process as it might apply to a 5 metric ton/day nuclear fuel reprocessing plant.

A pilot Combined Electrolysis Catalytic Exchange (CECE) system was designed and built using a hydrophobic precious metal catalyst developed by AECL at Chalk River and a solid polymer electrolyte electrolysis cell built by General Electric Company.

Accomplishments

The basic flow diagram and equipment descriptions for the CECE pilot scale system can be found in a previous report [1]. Figure 1 shows the system flow diagram used in the initial experiment. The two columns were connected to act as one long column with the tritiated water feed at the midpoint. As indicated in Figure 1, water feed to the top of the stripping section was distilled tap water. Hydrogen flow to the bottom of the enriching section was provided by electrolyzing water taken from the bottom of the enriching section. Since the basic catalytic exchange process concentrates tritium in the water at the bottom of the enriching section and lowers the tritium concentrations in the hydrogen at the top of the stripping section, a measurement of the effectiveness of the system can be obtained by the change in tritium concentration as a function of operating time at either point. In order to easily measure the hydrogen/tritium concentrations, a part of the depleted gas stream was oxidized. The resulting water was collected by bubbling it through ethylene glycol and was assayed for tritium via scintillation counting. These values are shown in Figure 1. At the end of the experiment, the water entering the electrolysis cells was similarly assayed, and this value is also shown in Figure 1.

Estimates of the equilibrium tritium concentrations at the two points are also shown in Figure 1. If a T:H separation factor of 7 is assumed, the equilibrium

FIGURE 1 - Combined electrolysis catalytic exchange system (CECE).

concentration of the hydrogen gas at the top of the stripping section is simply one seventh that of the water or 4×10^{-3} $\mu\text{Ci/liter}$. The concentration of tritium in the water at the bottom of the enriching section is not so easily calculated because, unlike the hydrogen streams at the top of the stripping section, the tritium concentration at this point is not known and is not constant. Using the parameters of this experiment, Butler [2] has estimated the equilibrium concentration of tritium to be $\sim 2 \text{ Ci/liter}$ at this point and the time to reach equilibrium to be $\sim 13 \text{ yr}$. The long equilibration time is primarily due to the large amount of tritium in the water contained in the electrolysis feed tanks. On the other hand, the hydrogen stream at the top of the stripping section reaches equilibrium relatively quickly because of the much higher volume flow rate, the lower total amounts of tritium in the hydrogen gas within the system, and the constant concentration of the water feed at this point.

The concentrations found in the equipment appear to confirm these estimates. It can be seen from the data in Figure 1 that the tritium concentration of the hydrogen stream always progresses toward the equilibrium value as operation time increases. This relatively rapid achievement of equilibrium in the stripped hydrogen stream is important since the primary purpose of the process is to strip tritium from the water.

The slow achievement of equilibrium in the water at the bottom of the enriching section does not pose a problem in a development system such as the one used in this experiment. However, in a production system it would probably be more convenient

to enrich to higher tritium concentrations and to achieve equilibrium more rapidly. This could be done by feeding water at higher tritium concentrations closer to the bottom of the enriching section and by reducing the water volume in the electrolysis section.

Probably more important than data obtained from this initial experiment with the CECE system is the insight into the operational problems involved in controlling the systems on a continuous basis for long periods of time. The system as conceived and built was to be controlled with feedback through a microprocessor. However, it was found that the controls were not capable of maintaining the necessary system parameters. It was necessary during this initial experiment to have constant supervision and control of the system parameters. The primary feedback controls are water level sensors in the bottom of each column. These sensors must keep the columns from flooding or running dry by adjusting the flow rates of water to the tops of the columns. Apparently the response time of the level sensors in the bottom of the column to a dry or too wet condition elsewhere in the column was not fast enough. In addition, all control activities in the system are hampered by the length of the lines between the water pumps and the columns. Each water stream from the bottom or to the top of a column must traverse more than 50 ft of 1/4-in. o.d. line.

Another control problem not connected to the microprocessor was the level control on the electrolysis feed tank. The level control failed to operate several times causing the cells to momentarily lose water flow. Quick manual operation of the fill pump prevented damage to the cells.

Eventual cause of the termination of the experiment was plugging of one of the valves in a water supply line. Apparently enough debris (mainly stainless steel wool and catalyst flakes) was present in the system to collect in and plug the narrow passages in the valve.

The first experiment using the CECE system was successful in demonstrating a reduction in tritium content of the hydrogen stream leaving the top of the catalyst column. Also demonstrated were problems in the microprocessor control of the system.

Future Plans

Problems involved with control of the system will be solved. The pilot CECE system will then be operated to obtain experience, determine scale-up parameters, and improve system dependability.

Electrolysis of high activity tritiated water

R. E. Ellis

Background

Operations involving tritium produce significant quantities of high activity (>10 Ci/liter) tritiated water. The tritium content of the hydrogen in such water is high enough to be economically used in gaseous processes for hydrogen isotope separations. However, in order to use the tritium, the water molecule must be separated into hydrogen and oxygen. Electrolysis is one of the more straightforward separation methods.

Results of a single experiment using a solid polymer electrolyte (SPE) electrol-

ysis cell were reported in an earlier document [3]. This experiment was successful in producing tritium from tritiated water, but was terminated because of catastrophic failure of the cell. From the experimental evidence it was not possible to determine if damage due to tritium caused the failure. Therefore, a new, larger capacity SPE cell was purchased for further tests.

Accomplishments

A system for electrolysis of high-activity tritiated water was designed, built, and tested. Figure 2 is a schematic for this system. With the exceptions of carbon gears in the pump and a small amount of Teflon in the flowmeter, the system is made of stainless steel to reduce corrosion. The active element is a General Electric SPE cell capable of electrical throughputs up to 50 A which corresponds to $18 \text{ cm}^3/\text{hr}$ of water.

In operation, water is brought into the oxygen-water reservoir through the pump. This water is pumped into the oxygen side of the cell where electrolysis takes place. Protons migrate through the SPE and become discharged producing hydrogen gas on the hydrogen side while oxygen gas is produced on the oxygen side. Both gases are saturated with water vapor, and liquid water is present on both sides of the cell. For these reasons, the oxygen and hydrogen reservoirs are equipped with spray traps to prevent entrainment of water in the gas streams. However, water is present in both gases at an equilibrium vapor pressure depending on the system temperature. If this loss of water proves to be a problem, cooling of the spray traps will be considered. From the reservoir the hydrogen gas goes to a storage tank and the oxygen is

FIGURE 2 - Electrolysis system schematic.

discharged to the effluent recovery systems (ERS).

The gas pressures are monitored by gauges at the top of each reservoir. GE recommends maintaining the pressure difference across the SPE at less than 100 psi. In practice it is planned to operate as close as possible to zero differential pressure in order to minimize damage in the event of SPE membrane rupture. Other monitors include a flowmeter and two level indicators. The flowmeter is a turbine type and is used to monitor the flow rate of the water circulating through the oxygen side of the cell. A flow rate of at least $50 \text{ cm}^3/\text{min}$ is necessary to remove waste heat from the cell when operating at maximum current.

The ultrasonic level indicators are non-contact devices located on the gas reservoirs and allow monitoring of the water level in each reservoir. On the hydrogen side this is necessary in order that the water be drained out of the reservoir and not allowed to overflow into the hydrogen storage system. On the oxygen side the level indicator is desirable so that the throughput of the cell can be estimated and the need for more feed water anticipated. Both the flowmeter and the level indicators are equipped with electronic as well as visual output so the system could be easily automated in the future.

The system as shown in Figure 2 has been successfully operated several times for periods up to 4 hr using deionized tap water. Current was varied during these experiments, but the cell did operate very satisfactorily at the maximum of 50 A. The level indicator on the oxygen side gave false "dry" indications at the higher

currents apparently from oxygen bubbles in the water. However, this false indication can be eliminated by momentarily turning off the power to the cell while a level determination is made.

A more serious problem is the large amount of water carried across the SPE to the hydrogen side. Apparently, each proton drags about four water molecules across the SPE. At 40 A this amounts to $60\text{--}70 \text{ cm}^3$ of water. The hydrogen reservoir will overflow in about 1 hr at this rate. Thus, every 40 min the system must be shut down, the hydrogen side must be evacuated, and the water must be drained from the hydrogen reservoir.

Future plans

The high activity tritiated water electrolysis system will be installed in the HTO synthesis glovebox. After initial checkout of the system, it will be used in an experimental investigation of electrolysis for recovery of tritium from water.

Fixation of aqueous tritiated waste in polymer impregnated concrete

J. J. Dauby, G. E. Gibbs, E. M. Pitre, and R. E. Wieneke

Introduction

Mound Laboratory, in cooperation with Brookhaven National Laboratory (BNL), has undertaken a program to test and develop the BNL method [4] for fixation of aqueous tritiated waste and adapt it to current procedures and facilities for packaging tritiated water waste. Concrete has been used at Mound since 1972 as a fixing agent for aqueous wastes containing 0.1 to 0.2 g of tritium per liter. In the

BNL polymer impregnation technique, the cured concrete is impregnated with styrene monomer. The monomer fills the pores; then polymerization of the monomer binds and immobilizes the radioactive waste in the concrete. This decreases the tritium release from the concrete and reduces contamination hazards at the burial site. Because the styrene permeates the concrete the waste is, in effect, "microencapsulated". This will significantly reduce the tritium release rate in the event the containment is breached. Thus incorporation of the BNL procedure can improve the present waste package and allow us to establish a lower tritium release rate for a waste burial container.

Small-scale tests were conducted to optimize parameters for polymerization of styrene in impregnated concrete. The tests samples consisted of 250 g of Type III Portland cement in a 16-oz polyethylene bottle with 25 wt % water added to the center of the sample through an injector at a rate of 2 ml/min. After the concrete cures at room temperature for five days in the polyethylene bottle, catalyzed styrene monomer is added and allowed to soak in. Polymerization was accomplished by holding the samples at 55°C for 24 hr. The results of the tests established an optimum amount of catalyst in the monomer at 0.5 wt % and an oven curing temperature of 55°C. The optimum weight ratio in a typical package remains under study, but samples to date favor a monomer-to-concrete ratio of 0.20.

Accomplishments

A test program was instituted to measure and compare the release of tritium from tritiated concrete with and without styrene impregnation. A 1000 cm³ tritiated water

solution containing 6.18 Ci/cm³ was prepared and assayed prior to use as a stock solution in preparing polymer-impregnated tritiated concrete samples. Sixteen hot samples containing tritium were prepared along with five blanks. All samples were prepared in 16-oz polyethylene bottles with a 2.625 in. (6.67 cm) diameter and filled to a height of 2.28 in. (5.79 cm). This height was chosen to maintain the same surface/volume ratio as was used in the 10 Ci samples prepared by BNL and sent to the Savannah River Plant for leach tests [5].

Approximately 62.5 ml of tritiated water containing 386 Ci was injected into each of the 13 samples containing 250 g of dry mix. This is six times the concentration of tritium that is in a normal tritiated waste package. Hot samples consisted of triplicate mixtures of cement-plaster in a 1:1 weight ratio, triplicate cement-plaster mixtures in a 1:1 volume ratio, and 10 samples of cement III. After the samples cured at room temperature for five days, catalyzed styrene monomer was added and they were placed in an oven. Some samples contain 5 wt % catalyst and were polymerized at 40°C. Most samples had 0.5 wt % catalyst and were polymerized at 55°C (see Table I).

The samples were put into a test program to measure the release rate of the tritium from the samples. The test method (shown in Figure 3) consists of submerging each sample, still contained in the polyethylene bottle, in 1600 cm³ of water which is sampled periodically to measure the tritium released from the sample.

The permeation data in Table I are given for the first six weeks of each sample

Table 1 - PERMEATION TESTS

Group	Sample No.	Curies	Preparation Date	Type	Water/Dry Mix Ratio	Styrene (%)	Catalyst (%)	Polymerization Temperature (°C)	Total Permeation (Ci)	Fractional Release
1	55	0	5-10-77	Cement III	0.250	20	5	40		
1	56	0	5-10-77	Cement III	0.250	20	5	40	None	
1	57	386.6	5-10-77	Cement III	0.250	20	5	40	0.40	1.02×10^{-3}
1	58	386.5	5-10-77	Cement III	0.250	20	5	40	0.37	0.97×10^{-3}
1	59	386.3	5-10-77	Cement III	0.250	20	5	40		
2	61	0	5-12-77	1:1 by weight	0.250	23	1/2	55	none	
2	62	386.1	5-13-77	1:1 by weight	0.250	23	1/2	55	0.42	1.08×10^{-3}
2	63	385.9	5-13-77	1:1 by weight	0.250	23	1/2	55	0.43	1.11×10^{-3}
2	64	386.6	5-13-77	1:1 by weight	0.250	23	1/2	55	0.48	1.25×10^{-3}
3	65	0	5-12-77	1:1 by volume	0.305	20	1/2	55	none	
3	66	386.6	5-13-77	1:1 by volume	0.305	20	1/2	55	0.40	1.03×10^{-3}
3	67	386.9	5-13-77	1:1 by volume	0.305	20	1/2	55	0.40	1.03×10^{-3}
3	68	386.6	5-13-77	1:1 by volume	0.305	20	1/2	55	0.59	1.52×10^{-3}
4	60	0	5-12-77	Cement III	0.250	20	1/2	55	none	
4	69	386.5	6-3-77	Cement III	0.250	20	1/2	55	0.50	1.30×10^{-3}
4	70	386.6	6-3-77	Cement III	0.250	20	1/2	55	0.55	1.42×10^{-3}
4	71	386.3	6-3-77	Cement III	0.250	20	1/2	55	0.24	0.63×10^{-3}
4	72	386.5	6-3-77	Cement III	0.250	20	1/2	55		
5	73	386.6	7-22-77	Cement III	0.250	none	none	none	0.37	0.95×10^{-3}
5	74	387.0	7-22-77	Cement III	0.250	none	none	none	0.47	1.21×10^{-3}
5	75	193.4	7-22-77	Cement III	0.250	none	none	none	0.35	1.81×10^{-3}

FIGURE 1 - Styrene impregnated tritiated concrete test.

FIGURE 4 - Fractional release from Cement III samples. Group 1.

FIGURE 5 - Fractional release from cement/plaster (1:1 by weight). Group 2.

FIGURE 6 - Fractional release from cement/plaster (1:1 by volume). Group 3.

FIGURE 7 - Fractional release from Cement III. Group 4.

even though most samples have been in the test for 16 weeks. The data were normalized to 6 weeks to facilitate comparison. It was difficult to draw any conclusions at this early date. However, preliminary results may indicate cement III retains the tritium of the sample better than either of the cement-plaster mixtures.

The data on samples 73 to 75 which are not impregnated with styrene do not yet give a clear indication. The data may indicate a higher release of tritium where styrene is absent as would be expected. Probably the polyethylene bottle is retaining the released tritium equally well for all the different sample types. The polyethylene bottle is thicker around the top and bottom, and the side walls are an average of 0.4 cm. The amount of polymerized styrene around the concrete samples is equivalent to 0.3 cm. Our conclusion is that the 386-Ci

FIGURE 8 - Fractional release from Cement III. Group 5.

tritium polymer-impregnated waste samples are showing about 0.4 Ci of tritium released in seven weeks of the test which is a fractional release of 1×10^{-3} from the samples in the polyethylene bottles.

Even after 17 weeks, little difference is observed among the test samples. For samples 57 and 58 (styrene-impregnated cement) an average of approximately 5×10^{-3} fractional release is observed. Samples 62 to 64 (styrene-impregnated 1:1 by weight cement-plaster) show an average fractional release of about 6×10^{-3} . Samples 66 to 68 (styrene-impregnated 1:1 by volume cement-plaster) show an average fractional release of about 5×10^{-3} , and the last set of styrene-impregnated cement (samples 69 to 71) show only about 4×10^{-3} fractional release of tritium.

The data of each sample in the test are shown in Figures 4 through 8. The blanks do not show an increase of tritium in the test water and the tritium is only just measurable above background. The blanks were prepared in the same contaminated facilities as the other samples and did have surface contamination of 500 to 2000 counts/min/40 cm² which was similar to the surface contamination of 2000 to 4000 counts/min/40 cm² of the tritiated samples

The first full-scale burial package (cold prototype) was prepared by the process developed by BNL and refined at Mound. Twenty-eight liters of water were added to a 27-gal polyethylene drum containing 112 kg of cement. The polyethylene drum was inside a 30-gal steel drum. After the cement cured at room temperature for nine days, 28 kg of styrene monomer with 140 g of catalyst was added to the polyethylene drum and allowed to stand for three days. It is not known how long it took for the styrene to soak into the concrete. The package was then heated to polymerize the styrene. The surface temperature of the steel drum was quickly brought to 90°C by band heaters which were then adjusted down to 60°C. The center-line temperature reached a maximum of 84°C after 16 hr. Upon examination of the package, it was noted that the styrene monomer had seriously damaged the polyethylene drum.

A second full-scale package was prepared using 112 kg of cement and 27 liters of water in the same manner as the first drum. After an 11-day cure at ambient temperature, the drum was gradually heated for 16 hr at which time the center temperature was 30°C. The 28 kg of styrene-monomer with catalyst was added with the

temperature held at 30°C. The absorption of the styrene monomer into the cement was completed after 4.5 hr at which time the band heaters were increased to 60°C. A maximum center temperature of 84°C was reached after 18 hr. Examination of the package showed reduced polyethylene damage. However a quantity of water escaped the cement and polystyrene during the polymerization process.

It has been determined that the small-scale laboratory tests had been conducted in high-density polyethylene containers whereas the 27-gal drum was a conventional low-density polyethylene. Therefore, an experiment was run to test the stability of both the low-density and the high-density linear polyethylene in styrene monomer. After 13.5 hr at 20°C plus 4.5 hr at 60°C, the low-density polyethylene was severely damaged, but the high-density linear polyethylene was only extended slightly. Further tests are under way to define the problem of corrosion of the 27-gal polyethylene drum by styrene monomer. It is anticipated that these tests will indicate how to lower the contact time between the drum and the styrene monomer and also indicate if there is a critical maximum temperature which should be avoided.

Future plans

The triplicate samples of cement, 1:1 ratio by weight of cement-plaster, and the 1:1 ratio by volume of cement plaster samples impregnated with styrene polymer will be checked periodically to measure the tritium released. Samples of cement III injected with 386 Ci of tritium without styrene polymer will also continue in the test program.

After the corrosion problems are overcome, full-scale 27-gal tests of polymer-impregnated concrete will resume in preparation for full-scale tritiated concrete tests.

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The authors would like to acknowledge H. Steinmeyer for synthesizing the tritiated water.

Management of high specific activity tritiated liquid wastes

G. E. Gibbs and J. J. Dauby

Background

Mound generates high-activity tritiated liquid waste from a variety of tritium handling operations and from stack effluent control systems [6-8]. There are two general categories of liquid waste which can be described as water (~ 1000 Ci/liter) and vacuum pump oil (~ 50 Ci/liter). Since 1972 these materials have been packaged for disposal in an improved container and the work has been done in a glovebox facility. Handling is accomplished with virtually no release to the environment and no significant operator exposure. The facility, package, and operating experience have been described in detail in the literature [9]. Waste oil is sorbed on vermiculite in the polyethylene drum but the remainder of the package is the same as for water. In both cases 27 liters of liquid is disposed of in each package.

Prior to the initial shipment of high specific activity waste to the Nevada Test Site, 96 drums of solidified liquid waste (water and oil) from our inventory were inspected (primary, secondary, and/or tertiary con-

tainer) to verify their condition and containment characteristics.

Of these, four had internal pressures of 10 psig in the polyethylene primary container. This gas was found to contain mainly hydrogen in addition to the argon and nitrogen originally present.

A study of tritium release from typical waste packages is continuing. To date, a maximum release of 1 Ci occurred from a package containing 90,000 Ci. The average daily fractional release from six packages is 2×10^{-8} .

Pressure tests were performed on both the components of the waste package and on the assembled package. The primary container can withstand a minimum of 30 psig internal pressure and 40 psig when contained in the secondary steel drum.

FIGURE 9 - Schematic of Waste Package

Accomplishments

DRUM RELEASE STUDY

A tritium release rate study of actual burial packages was initiated in May 1975 and is continuing. The study consists of the individual immersion of representative drums in a known volume of water. By periodic sampling of the surrounding water and analysis for the presence of tritium, a permeation rate can be calculated.

Figure 9 is a schematic of a typical burial package under test within the permeation apparatus. The next four figures, 10-13, are the resultant permeation rates (total permeation as a function of time in test) to date for the individual drums still in test. Pertinent information for each package is available on the graphs.

Since several variables remain unexplained, another graph, Figure 14 was prepared to compare the fractional release (total curies permeation ÷ total curies contained) as a function of the time since the package was prepared. This illustration takes into account the total curies actually in the package with consideration given to tritium decay as well as the time since the burial package was first assembled.

All drums ever in test are shown which lends itself to a comparison of the drums. As expected, the two oil drums (217 and 218) are almost identical and unexpectedly the fractional release is slightly higher than that from the water drums (205 and 318). Here, some consideration must be given to the fact that each drum is an individual and variations do occur. It should be noted that the more recent packages have lower fractional permeation.

Future plans

This test will continue and future results will be reported.

Radiation chemistry studies of tritium fixation packages

W. E. Tadlock and G. C. Abell

Background

Development of suitable containers for the disposal or storage of tritiated waste materials must be based on an adequate understanding of radiation-chemical effects caused by the tritium beta decay. Thus it is known [10] that gamma radiolysis of hydrocarbon oils generates appreciable quantities of hydrogen gas. The same is true for water; however, in a closed container the back reaction limits the pressure buildup [11]. Against this background, two questions about the suitability of a given containment package must be addressed: (1) what is the likelihood that an explosive mixture will develop and (2) what are the chances for container rupture due to pressure buildup?

Prior work

A previous report [12] outlines pressure buildup obtained on tritiated octane and tritiated water both with and without fixation. Gas composition of the overgas as well as $G(H_2)$ values are reported for the octane storage units.

Accomplishments

The pressure buildup measurements associated with tritiated octane (activity = 1000 Ci/liter) with and without fixation have been repeated using argon as the initial overpressure. In addition, for the octane

FIGURE 10 - Permeation rate for drum no. 217 containing 0.17 g (1632 Ci) of tritium in waste oil packaged on 10/1/74.

FIGURE 11 - Permeation rate for drum no. 218 containing 0.17 g (1632 Ci) of tritium in waste oil packaged on 10/1/74.

FIGURE 12 - Permeation rate for drum no. 318 containing 5.43 g (52128 Ci) of tritium in waste water packaged on 4/20/76.

FIGURE 13 - Permeation rate for drum no. 205 containing 3.80 g (36480 Ci) of tritium in waste water packaged on 7/74.

FIGURE 14 - Permeation rates for six drums.

FIGURE 15 - Pressure buildup over tritiated octane with and without fixation.

units without fixation; the measurements were repeated a third time using hydrogen as the initial overpressure. The purpose of this experiment was to determine whether the equilibrium hydrogen pressure was ≤ 35 psia which was the limit of the range of the pressure transducers. The pressure increase results which are shown in Figure 15 were essentially the same as those

obtained previously. Mass analysis of the overgas, Table 2, indicates that hydrogen constitutes about 85% of the gaseous product, a value that is comparable with previous results.

After a period of 1 yr, the pressure buildup for storage units containing waste vacuum pump oil (activity = 100 Ci/liter)

FIGURE 16 - Pressure buildup over tritiated water with and without fixation.

without fixation is only 3 torr. The pressure change for the units with vermiculite fixation is negligible with one unit having a +3 torr and the other a -8 torr change.

The units having water (activity = 1000 Ci/liter) with and without fixation show that

the rate of pressure increase has not changed. Figure 16 shows an updated graph of the pressure buildup data collected on the water storage units. In order to accelerate the results on the fixated water unit, the overgas was sampled and replaced with 20 psia hydrogen. The purpose here

is to determine whether the equilibrium hydrogen pressure is ≤ 20 psia. The $G(H_2)$ for the fixated water units was calculated to be 0.27 molecule/100 eV. Table 3 lists the mass analysis of the overgas from the storage units containing water with and without fixation. Storage times for the water without fixation was 153 days and for the fixated water was 573 days. The concentration of nitrogen, which could have come only from air in the overgas, suggests that about 0.8% oxygen was depleted probably by scavenging of radicals.

Future plans

Storage units having tritiated water contained in polymer-impregnated concrete are being prepared. Pressure buildup measurements will follow.

Table 2 - MASS SPECTROMETRIC ANALYSIS OF OVERGAS FROM OCTANE STORAGE UNITS

CONSTITUENT	Without Fixation (mole %)	Over Vermiculite (mole %)
H ₂	42.5	46.7
Hydrocarbons	4.5	4.8
A	49.7	43.9
N ₂	2.7	3.4

Table 3 - MASS SPECTROMETRIC ANALYSIS OF OVERGAS FROM WATER STORAGE UNITS

CONSTITUENT	Without Fixation (mole %)	Over Cement Plaster (mole %)
H ₂	15.9	37.4
N ₂	3.5	3.5
O ₂	0.00	0.0
A, He	78.2	59.0

Cryogenic distillation

W. R. Wilkes

Background

The most economical method of enriching hydrogen isotopes to high concentrations, e.g. more than a few percent of deuterium or tritium, is by means of cryogenic distillation. The reason is that the vapor pressure ratios of the isotopes are large, of the order of 1.5 to 3, and the isotopes are much more dense in the liquid state than in the gaseous state required for another common separation method, gaseous thermal diffusion. Thus, large amounts of space and high electrical power inputs, as are required for thermal diffusion, can be avoided in a distillation system.

A cryogenic distillation column was built and operated a few years ago to study hydrogen isotope distillation. This column was constructed in a laboratory which is unsuitable for tritium work, so a new column was constructed in the Tritium Effluent Control Laboratory.

Accomplishments

The cryogenic distillation apparatus which had been installed earlier in the Tritium Effluent Control Laboratory was leak checked and some minor modifications and repairs were made to bring the system into operating condition.

No on-line facilities were provided for hydrogen isotope analysis, so a gas chromatograph was installed in the glovebox beside the distillation apparatus. The chromatograph is quite similar to others used at Mound for hydrogen isotope analysis. It utilizes a matched pair of 3 mm (1/8 in.) o.d. copper columns, 3 m in length which

are kept at 77 K by a liquid nitrogen bath. The columns, which are packed with 80-100 mesh alumina, are partially deactivated with oxygen gas. The oxygen, being paramagnetic, serves a dual function; it promotes the molecular conversion reaction (e.g., $H_2 + D_2 \leftrightarrow 2HD$) as well as filling many of the absorption sites in the alumina. The conversion is necessary so that ortho- and para- hydrogen (or deuterium or tritium) appear at the column output as one peak rather than as two. The site filling permits elution of a sample in minutes rather than hours. The carrier gas used is neon, which provides greater sensitivity for hydrogen than does helium.

The distillation system is similar in design to one constructed earlier in another building, but there are some noteworthy modifications.

First, the new system incorporates electromechanical flow control valves. These valves are operated by a signal obtained from a mass flowmeter which is integral to the valve. The use of these valves permits a constant flow rate to be maintained even with pressure fluctuations within the process lines.

Each of the flow controllers can be set manually to the desired flow, and in addition two of the controllers may have their set points determined by column operating conditions.

Another improvement is the use of a metal bellows circulating pump instead of small, magnetically driven vane pumps. The vane pumps used earlier did not provide enough pressure differential to allow the desired mass flow rates.

The distillation column is similar in dimension to the earlier one, but this time has been constructed as three discrete units. The three units, boiler, condenser, and packed column, are each fabricated with commercial metal gasket fittings (Cajon VCR) at their connection points. This method of construction permits any one of the units to be replaced without using any tools other than a pair of wrenches. As this new column is to be operated in a helium filled glovebox, it is particularly important that neither soldering nor welding is required. The metal gasket seals have been made up repeatedly with no sign of leakage, and they have been thermally cycled to below 10 K several times, again without leaking.

The initial tests of the distillation system were with a packed column, 0.6 cm inside diameter and 10 cm long. The packing is 0.3 cm x 0.3 cm stainless steel Eglin packing. The gas used in the tests was either pure hydrogen or a mixture with a composition of approximately 81% H_2 , 18% HD, and 1% D_2 .

An attempt was made to measure the maximum permissible boilup rate as a function of column pressure. This was not possible, however, because the vapor velocity required to flood the column is so high that the capacity of the refrigerator was exceeded before a limit could be found. The measurements were made with hydrogen gas. In order to try to obtain a limiting vapor velocity, the column pressure was lowered to as little as 300 torr. This resulted in an increased vapor velocity without lowering the temperature enough to significantly reduce the refrigeration capacity. At this pressure, there was no sign of loss of liquid in the boiler at a vapor velocity of 20 cm/sec.

At a later time this measurement will be repeated with either D₂ or HD in the column. The lower vapor pressure of these liquids will allow the extension of these tests to higher velocities.

The height of a single separation stage was measured using a mixture of H₂, HD, and D₂. Stage heights were measured at 760, 1000, and 1500 torr. The stage heights were measured at various boiling rates but with constant product withdrawal rates. The withdrawal rates were small enough to permit reflux ratios of the order of 10 to 100. There was so little deuterium in the separation system that even in the boiler, where the deuterium concentration is the highest, there was less than 10% deuterium. For this reason, a rough estimate of the number of stages, N, was made using the Fenske equation,

$$N + 1 = \ln \frac{XH_2(\text{Top}) XHD(\text{Bot})}{XHD(\text{Top}) XH_2(\text{Bot})} / \ln \alpha$$

and ignoring the deuterium fraction in the boiler. Here the X's are the mole fractions of hydrogen or hydrogen deutride (ignoring deuterium) in the top or bottom product, and α is the average of the ratio of the vapor pressures at the warm and cold ends of the column.

Stage height values of $H = 10 \text{ cm}/N$ measured this way ranged from 1.35 to 2.1 cm. There was a direct relation between boilup rate and stage height, with the lowest stage heights occurring at the lowest boilup rates. At even the highest boilup rates measured, 1.8 W in the reboiler, the stage height value of $\sim 2 \text{ cm}$ was substantially smaller than measured at LASL, where values of 5 cm have been reported [13].

Future plans

Tests of maximum tolerable vapor flow will be conducted using HD and/or D₂. Stage height tests will be concluded, and then a larger column $\sim 50 \text{ cm}$ will be installed. Tests will be performed to verify that good separations are obtained with H₂/HD mixtures in the larger column. Then H₂/HT mixtures will be introduced. Concentrations of HT will initially be about 1-10 ppm at the feed point.

Tritium containment systems

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Background

The Glovebox Atmosphere Detritiation System (GADS) is based on a helium flow loop in which air contaminants and hydrogen, including tritium, are removed to levels below 1 ppm by adsorption on a fixed bed of molecular sieve pellets cooled by liquid nitrogen. Water vapor is removed in a precooling, gas-to-gas heat exchanger and is collected in a liquid holding tank during the regeneration by heating to 120°C while purging with helium. The GADS was fabricated by C.V.I. Corporation of Columbus, Ohio. The system has a nominal flow design of 3 m³/min with dual column operations. Each adsorber column contains 300 lb of Linde 5A molecular sieve. Each column is on stream for 24 hr while the other is in regeneration mode. The columns operate near atmospheric pressure at 77 K when on stream and are regenerated by heating at 120°C with purge. The system was designed to maintain outlet impurity levels of less than 1 ppm for oxygen, nitrogen, water, carbon dioxide, hydrocarbons, and hydrogen.

In particular, the sum of all hydrogen isotopes is not to exceed 0.01 ppm at the outlet. The system was designed to remove from the atmosphere and contain 1 g of hydrogen (tritium) in the event of a sudden release in the glovebox. Such a release would raise the initial concentration in the inlet helium atmosphere to 465 ppm.

The Emergency Containment System (ECS) is an automatically actuated, room-air-detritiation system utilizing a catalytic oxidation reactor and presaturated oxide adsorption/exchange columns. In the event of an accidental release of tritium to the laboratory, the ECS is automatically activated and the quick-acting pneumatic dampers divert the room-air supply and exhaust through the ECS until the tritium concentrations in the room air are returned to safe operating levels. The air stream is heated to a temperature of 175°C before it enters the catalyst to meet the design goal of 99.9% oxidation of tritium in air at inlet concentrations of 0.5 ppm. The air is cooled to near 20°C before entering the adsorption columns. The oxidized tritium is captured and contained on the adsorption columns in the oxide form. The ECS is designed to provide the oxidation and adsorption capacities for a single pass decontamination factor of 1000:1 in a 30 m³/min air flow containing as high as 1 Ci/m³ tritium and 0.5 ppm natural hydrogen background. The adsorber section of the ECS consists of two stainless steel vessels containing 3800 lb of Alcoa alumina H151 adsorbent, saturated at 100% relative humidity. These vessels were sized to provide 10 hr of operation with an inlet activity of 1 Ci/m³ before a defined breakthrough of 100 µCi/m³ occurs. A Spencer turbine controls gas flow for the ECS.

Accomplishments

ECS EXPERIMENTS

The efficiency of the ECS to capture tritium released into an operating area was evaluated by releasing four elemental tritium samples into the laboratory. The automatic startup of the ECS was controlled by a 20-liter ionization chamber monitoring the laboratory. The exhaust from the area was also monitored by a 20-liter ionization chamber. Bubbler systems containing ethylene glycol sampled the laboratory exhaust, the area adjacent to the exterior doors, and the supply to the room coming from the ECS adsorber beds [14]. These systems provided data on the quantity of tritium oxide at the various sampling locations. Fifteen 100-cm² areas at various locations throughout the laboratory were marked off to determine tritium deposition on laboratory surfaces. Tritium deposition was determined by wiping the areas after the tritium release with dry Metrical filter paper. The wipes were counted by liquid scintillation counting.

After each experiment the adsorber bed used was regenerated by purging it with an air stream heated to 100°C. An ethylene glycol bubbler system sampled the effluent air stream to determine the quantity of tritium captured on the bed.

First Experiment

In the first experiment 0.86 Ci of elemental tritium was released followed by 5 liters of hydrogen. The hydrogen stream was diluted with a 0.03 m³/min flow of nitrogen to maintain a hydrogen concentration of less than 50% of the lower explosive limit of 4%. The 5 liters of hydrogen released into the 560 m³ room raised the hydrogen concentration to approximately 10 ppm,

20 times the design criterion of 0.5 ppm. tritium at the catalytic reactor inlet.

Real-time laboratory tritium concentration data were not obtained. A valve on the sampling system failed and prevented a continuous sample from reaching the ionization chamber. The exhaust monitoring system was operational, and at a concentration of $770 \mu\text{Ci}/\text{m}^3$ the ECS was manually activated. This occurred approximately 1.5 min after the release. Figure 17 shows the effluent levels during the experiment. The total quantity of the tritium lost through the exhaust system was 0.41 Ci or 47% of the quantity released. Eighty-three percent of this amount, or 0.34 Ci, was apparently beyond the exhaust shut-off dampers before the ECS was activated. The remaining 0.07 Ci was released as a result of leakage through the dampers. Although the dampers are quick acting, they are standard air dampers with an estimated leakage of 10%. Regeneration of the adsorber bed used for this experiment resulted in 0.27 Ci being removed. This corresponds to 31% of the quantity released. The total quantity of tritium accounted for was 79% of the 0.86 Ci released. The estimated error in the tritium content of the samples released is $\pm 15\%$. All wipe areas showed no detectable quantities of tritium.

The average tritium oxide concentration in the laboratory during the experiment was $1.8 \mu\text{Ci}/\text{m}^3$ which is 36% of the Radiation Concentration Guide (RCG) for occupational exposure. The tritium leakage through the doors of the laboratory was determined by the bubblers and an ionization chamber tritium monitor. The bubblers indicated an average tritium oxide concentration of $0.01 \mu\text{Ci}/\text{m}^3$. The tritium monitor indicated nondetectable concentrations of elemental

and tritium oxide. During the 1-hr experiment 0.003 Ci of tritium oxide was released through the ventilation system. This indicates that less than 1% of the tritium released was in the oxide form. Bubbler data from sampling the exhaust from the adsorber bed supply to the laboratory indicate 0.009 Ci of tritium was released back into the laboratory. The 0.009 Ci recirculated is 3% of the 0.27 Ci captured on the bed during the experiment. Thus the system was capable of capturing 97% of the tritium oxide produced by the system.

Second Experiment

In the second experiment 0.87 Ci of elemental tritium was released with 20 cm^3 of hydrogen. A nitrogen purge was used to flush the container to ensure all the tritium was released. The 20 cm^3 of hydrogen increased the hydrogen concentration to approximately 0.03 ppm above the natural abundance of 0.5 ppm.

Figure 18 is a plot of the tritium concentration in the laboratory resulting from the second experiment. The ECS was automatically activated approximately 10 sec after the sample was released. Figure 19 shows the effluent levels during the experiment. During the first 2 min of the experiment 0.08 Ci passed the dampers before the ECS was activated. An additional 0.28 Ci which was released during the experiment is attributed to leakage through the dampers. The total quantity lost to the exhaust system was 0.36 Ci or 41% of the quantity released. Regeneration of the adsorber bed resulted in the removal of 0.37 Ci or 42% of the material released. The tritium oxide concentration in the laboratory increased during the experiment to $2.9 \mu\text{Ci}/\text{m}^3$ or 60% of the RCG. Tritium oxide concentration exterior to the labora-

FIGURE 17 - ECS Experiment 1; exhaust system concentration as a function of time.

FIGURE 18 - ECS Experiment 2; laboratory concentration as a function of time.

FIGURE 19 - ECS Experiment 2; exhaust system concentration as a function of time.

tory was less than $7 \times 10^{-3} \mu\text{Ci}/\text{m}^3$. During the experiment, 0.003 Ci of tritium oxide, which is less than 1% of the total quantity released, escaped through the ventilation system. Sampling of the exhaust leaving the adsorber bed indicated that 0.006 Ci was released back into the laboratory. This would indicate that of the 0.37 Ci captured on the bed during the 70-min experiment 99% of the tritium processed by the ECS was captured on one pass. Approximately 10 min were required before the air stream reached 175°C.

Third Experiment

The third experiment duplicated the first experiment with the exception that the preheater was shut off to prevent the air stream from reaching the 175°C design temperature. The only heat supplied to the system was from the heat of compression from the turbine servicing the ECS.

The purpose of this experiment was to determine the efficiency of the system without preheating the air stream to 175°C. The air stream did reach a temperature of 80°C due to the heat of compression from the air turbine. Figure 20 shows the laboratory concentration of tritium. The ECS was automatically activated approximately 20 sec after the release. This experiment required 120 min to reduce the tritium concentration to less than $5 \mu\text{Ci}/\text{m}^3$ as compared with the 60-70 min required for the previous experiments. Figure 21 shows the effluent concentrations of tritium during the experiment. During the first 3 min 0.15 Ci was released. An additional 0.39 Ci was released during the remaining time. The total tritium lost to the exhaust system was 0.54 Ci or 61% of the quantity released. Regeneration of the adsorber bed removed 0.61 Ci or 69% of the

release. Tritium oxide concentration in the laboratory averaged $3.5 \mu\text{Ci}/\text{m}^3$ during the experiment. Oxide concentration exterior to the laboratory averaged $0.61 \mu\text{Ci}/\text{m}^3$. The total quantity of tritium oxide release through the exhaust system was 0.024 Ci or 4% of the total. The total tritium exhausted from the adsorber bed was 0.007 Ci during the experiment. Of the 0.61 Ci captured on the bed, less than 1% was recycled into the laboratory.

Table 4 show the tritium deposition on laboratory surfaces for the first three experiments. The deposition on the liquid nitrogen dewar during the third experiment indicates a significant increase in surface contamination on surfaces at low temperatures. It is apparent from the results of the tritium deposition tests that large quantities of tritium are not deposited during the brief period required to decontaminate the laboratory air.

Substantially higher surface concentrations were found after experiment 3 than after experiments 1 and 2. This can be explained qualitatively by the fact that high airborne concentrations persisted for longer times than in the earlier experiments.

Fourth Experiment

In the fourth ECS experiment 0.88 Ci of elemental tritium was released with no additional hydrogen. Prior to the release, the laboratory supply and exhaust were shut down and weather balloons were inflated inside the exhaust and supply ducts. These actions were taken because the first three experiments showed that the dampers leaked thus releasing tritium through the exhaust duct. The balloons provided a protective seal. Prior to the tritium release, the ECS was manually activated to prevent any tritium loss during the

FIGURE 20 - ECS Experiment 3; laboratory concentration as a function of time.

FIGURE 21 - ECS Experiment 3; exhaust system concentration as a function of time.

Table 4 - TRITIUM DEPOSITION ON LABORATORY SURFACES

Surface	Tritium Deposited (dis/min/100 cm ²)		
	Experiment 1	Experiment 2	Experiment 3
Iron	Nondetectable	Nondetectable	48
Sheet Metal	Nondetectable	Nondetectable	26
Plexiglas	Nondetectable	Nondetectable	55
Glass	Nondetectable	Nondetectable	35
Bench Top	Nondetectable	Nondetectable	33
Glass	Nondetectable	Nondetectable	19
Desk	Nondetectable	9	21
Bench Top	Nondetectable	Nondetectable	84
Sheet Metal	Nondetectable	Nondetectable	21
Glass	Nondetectable	5	31
Wood	Nondetectable	Nondetectable	36
Tile	Nondetectable	2	22
Tile	Nondetectable	Nondetectable	36
Desk Top	Nondetectable	Nondetectable	21
Tile	Nondetectable	Nondetectable	19
Liquid Nitrogen Dewar			3388
Ice on Liquid Nitrogen Dewar			26704

time required for the monitoring systems to activate the ECS. This experiment was performed to simulate a system which would have low leakage dampers located at a sufficient distance downstream to prevent any tritium from passing the dampers before they closed.

Figure 22 shows the laboratory concentration of tritium during the fourth ECS experiment. The exhaust monitoring system level increased to $2 \mu\text{Ci}/\text{m}^3$ after 10 min and returned to background in 10 min. There was no measurable air flow at the sampling location, and, therefore, the total quantity of tritium released was insignificant. The average tritium oxide concentration in the laboratory was $0.52 \mu\text{Ci}/\text{m}^3$. The bubbler system monitoring the adsorber bed exhaust indicated that a total of 0.005 Ci was recirculated into the laboratory. Regeneration of the adsorber bed accounted for 0.59 Ci or 66% of the quantity released. The total quantity of tritium oxide recirculated into the laboratory was 0.8% of that processed by the system. The 15 wipe areas showed surface contamination levels of 72-137 dis/min/100 cm^2 . Tritium monitors exterior to the laboratory showed no detectable concentrations of tritium.

Data Analysis

The concentration data obtained during the last three ECS experiments were used to determine the efficiency and decontamination factor for the ECS. If a well-mixed atmosphere is assumed, the fraction of tritium remaining in the room can be expressed by Equation 1 and the fraction released to the environment can be expressed by Equation 2.

$$T = e^{-(Y_L + \epsilon Y_R)t} \quad (1)$$

$$T' = \frac{Y_L}{Y_R + Y_L} [1 - e^{-(Y_R + Y_L)t}] \quad (2)$$

where T = the fraction of the tritium initially present that remains in the room after time t .

T' = the fraction of the tritium initially present that is leaked to the environment in time t .

t = time, hr.

Y_R = removal time constant, $\text{hr}^{-1} = 4.2$.

Y_L = leak time constant, hr^{-1} . This is not known directly, but may be determined from the values of T and T' .

ϵ = system tritium removal efficiency. This too must be calculated from T and T' .

The decontamination factor (DF) is defined as the reciprocal of the fraction of tritium initially present which remains uncontained after time t .

$$DF = \frac{1}{T + T'} \quad (3)$$

After 25 min during experiment 2, the entire area serviced by the ECS was at a uniform concentration of $110 \mu\text{Ci}/\text{m}^3$ as determined by the fact that two air monitors at different parts of the laboratory showed the same concentration. The total quantity of tritium in the 560-m^3 area at that time was 0.06 Ci. The concentration then decreased to 0.009 of the original or $1 \mu\text{Ci}/\text{m}^3$ in 0.75 hr. The quantity released to the effluent during this time period was 0.01 Ci or 0.17% of the initial quantity of tritium in the area.

Substituting into Equation (1) and (2), one finds that the calculated efficiency

FIGURE 22 - ECS Experiment 4; Laboratory concentration as a function of time.

of the ECS is 1.4. An efficiency greater than one is presumably the result of incomplete mixing; i.e., the tritium concentration in the laboratory was not uniform.

The decontamination factor for experiment 1 was calculated to be 6.

A uniform concentration of $38 \mu\text{Ci}/\text{m}^3$ was assumed after 60 min during experiment 3. It required 1 hr more to decrease the concentration to $1 \mu\text{Ci}/\text{m}^3$. During this period 0.003 Ci of the 0.021 Ci in the room at the beginning of the period was released to the ventilation system. The calculated efficiency of the ECS system was 0.7. The calculated efficiency of the catalysts at 80°C is 0.9 [15]. The decontamination factor during experiment 2 was determined to be 6.

The equilibrium concentration during the fourth ECS experiment occurred in 25 min and was $266 \mu\text{Ci}/\text{m}^3$. After 1.6 hr the concentration was $1 \mu\text{Ci}/\text{m}^3$ or 0.003 of the original concentration. No measurable quantity of tritium was released to the ventilation system. The efficiency of the system was determined to be 0.9. The decontamination factor during the 1.6 hr was 333. The decontamination factor for the entire 2-hr experiment was 1800. The predicted decontamination factor using the model was 1900. Note here that the decontamination factor increases with time. In experiments 2 and 3, because of leakage, the decontamination factor approaches a limiting value after long cleanup times.

The fourth ECS experiment has shown that the ECS will function as designed if low leakage dampers are installed.

Of the 2.2 Ci captured on the adsorber beds, 1.8 Ci were accounted for through regeneration of the beds. The unaccounted for material is assumed to be held on the 3500 kg of adsorber. Samples of the adsorber material will be taken to determine what quantity of tritium still remains on the adsorber.

GADS EXPERIMENT

The Glovebox Atmosphere Detritiation System (GADS) was evaluated by releasing two tritium standards into a 2-m^3 glovebox serviced by the system. One standard contained 0.64 Ci of elemental tritium in 911 cm^3 of hydrogen. The additional hydrogen released increased the hydrogen concentration in the glovebox to 500 ppm in order to simulate the design criterion of 465 ppm. The sample container was flushed with helium to ensure that all the tritium was released. Oxygen concentration in the glovebox was less than 1 ppm prior to the tritium release.

The second standard contained 0.86 Ci of elemental tritium in 20 cm^3 of hydrogen. This standard was also flushed with helium. This experiment was conducted to determine the efficiency of the GADS at hydrogen concentrations of less than 2% of the design criterion. Oxygen concentration in the glovebox was 25 ppm prior to this release.

Wipes of the interior of the glovebox were taken after each of the two release experiments. A 100-cm^2 area was wiped using dry Metrical filter paper, which was then analyzed by liquid scintillation counting.

The inlet tritium concentration to the GADS was monitored with a 2-liter ionization chamber utilizing a vibrating reed electrometer.

The GADS reduced the tritium fraction in the glovebox during the first experiment to 0.0006 in 17 min. From Equation 1 the efficiency of the system was calculated to be 0.56. During the second experiment the GADS reduced the tritium fraction to 0.0004 in 13 min. The calculated efficiency for the experiment was 0.77. Figures 23 and 24 show the concentration in the glovebox for experiments 1 and 2 respectively. Wipes of equipment inside the glovebox, including stainless steel, iron, Plexiglas, and rubber, ranged from 1051 to 1575 dis/min/100 cm². The results of these experiments indicate that the GADS did not function to the design goal of 0.99998. The model assumes that there is one complete

air turnover in the glovebox for each equivalent air pass. The physical layout of the glovebox exhaust and supply would indicate that a 100% dilution is not readily achievable. The GADS is therefore anticipated to have a tritium removal efficiency greater than those calculated.

Future plans

A fast-acting and low-leakage damper system will be installed in the ventilation system in the areas served by the ECS. Additional releases will then be made to determine the efficiency of the ECS. Data on tritium deposition on various materials under different environmental conditions will also be collected.

Modifications will be made to the glovebox serviced by the GADS, and additional tritium releases will be made and evaluated.

FIGURE 23 - GADS Experiment 1; inlet concentration as a function of time.

FIGURE 24 - GADS Experiment 2; inlet concentration as a function of time.

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