MLM-2087(OP)

NEW APPROACH TO TRITIUM LEAK RATE DETERMINATION

Conf-731112--8

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Summary

A method for determining leak rates (on the order of 1 x 10^{-17} cm^3/sec and higher) from containers of tritium waste is described. Each container is sealed in a disposable can. After several days, the accumulated tritium is flushed from the can with compressed air through an ethylene-glycol bubbler where the HTO, but not the HT, is trapped. The output of the bubbler is channeled through a heated palladium catalyst where the HT is oxidized. The oxidized HT is collected in a second ethylene-glycol bubbler. The tritium collected in both bubblers is measured by liquid scintillation counting.

Introduction

A method for determining tritium leak rates was described by one of the authors at the Las Vegas Tritium Symposium in 1971.[1] The measurements are required in support of the Mound Laboratory program for recovery of tritium from waste materials. The techniques are used to monitor containers of tritiated materials for leakage and to check joints and seals of parts as used in the recovery system, part of which is operated at 3 atm pressure. Tritium is difficult to contain, and it is important that there be no release to the environment. Tritium in the form of tritiated water (HTO) is more hazardous in the environment[2] than tritium gas (HT). The leak test procedures were therefore improved to increase the sensitivity, to distinguish between HT and HTO, and to decrease the manhours required per measurement.

Collecting and Measuring the Tritium Leakage

The container of tritiated material or part as used in the recovery system, is sealed in a collection chamber, where the tritium is allowed to accumulate for several days. Inexpensive cans are used for collection chambers. One and five gallon tin cans (Freund Can Co., Chicago, Ill.) and 16 gallon cans (Heekin Can Co., Cincinnati, Ohio) are shown in Figure 1. A "quick-connect" Swagelok coupling is attached to each side of the can. The one gallon can is mechanically sealed. The seams of the larger cans are soldered and, with the aid of sealing putty, are made airtight.

The outside of the part or container is wiped free of dust and grease and then is sealed in an appropriate size can (collection chamber). Since surface contamination unduly influences apparent leakage as measured over short periods of time, room air is pumped through the can for 16 hr or more. For the purposes of this test, this effectively decontaminates the surface of the container sealed inside. The valves on the can are then closed, and the container is allowed to remain sealed inside the can for four days or longer.

After this static period, part of the tritium inside the can is in the atmosphere of the can, and part of it may have adsorbed on the sides of the can and on the surface of the container or part whose leak rate is being measured. A significant fraction of the tritium will be adsorbed if it is in the form of HTO. The measured leak rate must include the adsorbed tritium and the tritium in the can atmosphere.

In the procedures described at the Tritium Symposium,[1] the interior of the can was coated with cellulose acetate before sealing. After a static period the coating was peeled off and dissolved in scintillator solution, and the adsorbed tritium was measured by liquid scintillation counting. Aliquots of the atmosphere were measured by proportional counting. These procedures were time consuming. The sensitivity reported was 1 x 10^{-16} cm^3/sec.

New Approach

With the new approach, the cellulose acetate coating is eliminated. After the static period, the accumulated tritium (both the adsorbed and atmospheric) is flushed from the can and measured. The method used to recover and measure the tritium is a modification of the palladium catalyst oxidation technique developed at Mound Laboratory for monitoring stack gas exhaust.[3-5] Figure 2 is a diagram of the leak detection system. Compressed air is directed through a heated palladium catalyst to oxidize any HT which may be in the air stream, and then through a Drierite trap to remove the HTO. The resulting, dry, tritium-free air is then channeled through the can (collection chamber), flushing the collected tritium from the can through an ethylene-glycol bubbler where the HTO, but not the HT, is trapped. The output of the bubbler is channeled through a second palladium catalyst heated to 400°C, where the HT is oxidized, and into a second bubbler, where the oxidized HT is trapped. Thus the HTO which had accumulated in the can is collected in the first bubbler and the HT in the second.

The bubbler is shown in Figure 3. It is a conventional 25-ml polyethylene liquid scintillation vial containing 10 ml of ethylene glycol. Air is flushed through stainless steel tubing at a rate of 750 cm^3/min into the glycol where the moisture is collected. It is vented through another piece of tubing through a specially made cap. The cap remains in place on the system. The vial of glycol is

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under U. S. Government Contract No. AT-33-1-GEN-53.

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removed for counting and replaced with another vial.

Prolonged flushing is required to desorb and remove all of the tritium from the inner surface of the can and from the surface of the container or parts. The fraction of tritium adsorbed is a function of the amount of HTO compared to total tritium and the duration of the static interval. A succession of flush periods are used for each leak test, varying in duration from 1 to 16 hr, depending upon the size of the can and the convenience of the analyst. At the end of each flush period, the trap is removed and replaced with another vial of glycol. The exposed glycol is mixed with scintillator solution and measured for tritium content by liquid scintillation counting.

Computation of Leak Rate

It has been established that the leak rate into static moist atmosphere (room air) is not equal to the leak rate when dry air is being flushed through the can. The total tritium collected in dis/min (T) in both bubblers is:

$$T = r_s t_s + r_f t_f \qquad (1)$$

where r_s and r_f are the static and flush leak rates in dis/min/sec, and t_s and t_f are the static and flush time intervals in seconds. The static leak rate, r_s, cannot be measured directly, but is given by the following, derived from equation 1:

$$r_s = \frac{T - r_f t_f}{t_s} . \qquad (2)$$

Several days flushing may be required to remove all the adsorbed tritium. After this is accomplished, a constant leak rate, r_f, is observed for two or more flush intervals.

The leak rate (L) in cm^3/sec into static room atmosphere is given by:

$$L = \frac{r_s}{5.26 \times 10^{12}} \qquad (3)$$

where 5.26×10^{12} is the disintegration rate in dis/min of 1 cm^3 of tritium at 1 atm pressure, 25°C.

Since the first bubbler traps the tritium which leaves the can as HTO and the second bubbler collects the oxidized HT, the fraction of the total leak rate which is in the form of the oxide can be calculated.

Leak rates as low as 1×10^{-17} cm^3/sec can be measured by these techniques. A 20-day static collection period is desirable at this level.

An example of the data and computation of a leak rate determination is shown in Table 1. The container of tritium was sealed in ambient atmosphere and allowed to stand for five days. Dry air was then flushed through the can as described. During the first flush period of 4 hr duration at 750 cm^3/min, the atmosphere inside the can was displaced with dry air, taking the tritium in the air with it. An examination of the data will show that an additional 24 hr were required to remove most of the adsorbed tritium. A steady leak rate, r_f, was observed during the last two flush

Table 1

LEAK TEST DATA AND COMPUTATIONS

Static Time - 5 days, 4 hr, 20 min

Flush Interval	Flush Time (hr)	HTO in Bubbler #1 (dis/min)	HT in Bubbler #2 (dis/min)	Leak Rate Per Flush Interval (dis/min/sec)
1	4	120,165	977	---
2	16	37,838	888	0.672
3	8	12,309	325	0.439
4	16	19,267	594	0.345
5	3	3,712	83	0.351
Totals	47	193,291	2,867	

Computations

$T = 196{,}158$ dis/min

$t_s = 4.476 \times 10^5$ sec

$t_f = 1.692 \times 10^5$ sec

$r_f = 0.346$ dis/min/sec*

$$r_s = \frac{T - t_f r_f}{t_s} = 0.3075 \text{ dis/min/sec}$$

$$L = \frac{r_s}{5.26 \times 10^{12}} = 5.8 \times 10^{-14} \text{ cm}^3\text{/sec}$$

*Weighted average for last two flush intervals.

HTO $\geq$ 98.5% of total tritium.

intervals. It will be noted that 98.5% of the tritium leakage in this test was in the form of HTO. It is probable that at least part of the tritium found in the second bubbler is HTO carried over from the first bubbler. This could be avoided by inserting another bubbler between the HTO and HT traps.

Validity of Procedures

The procedures have been tested by introducing known amounts of HT and HTO into cans, allowing the cans to remain sealed for several days, and then proceeding as for a routine leak test. The HT introduced into the cans was first standardized by proportional counting and the HTO by liquid scintillation counting. The accuracy of both is directly traceable to NBS standards.[1] Results given in Table 2 indicate the procedures are sound.

The manhours required for a leak rate measurement is roughly 1/10 of that required using the former procedures.

Acknowledgement

The authors acknowledge with gratitude the assistance of R. L. Ryan in the development of these procedures.

References

1. M. L. Curtis, C. T. Bishop, M. M. Bolton, and R. L. Ryan, "Determination of Tritium Leak Rates from Waste Processing Equipment," in Tritium, A. A. Moghissi and M. W. Carter (ed.), Messenger Graphics, Las Vegas, Nevada, 1973, pp. 784-791.
2. D. G. Jacobs (ed.), Sources of Tritium and Its Behavior Upon Release to the Environment, TID-24635 (1968), p. 1.
3. D. C. Carter et al., Mound Laboratory Chemistry and Physics Report: October-December 1971, MLM-1888 (August 21, 1972), p. 19.
4. D. C. Carter, Mound Laboratory Chemistry and Physics Report: January-March 1972, MLM-1903 (October 20, 1972), p. 22.
5. D. C. Carter, Mound Laboratory Chemistry and Physics Report: April-June 1972, MLM-1955 (December 29, 1972), p. 28.

Table 2

VERIFICATION OF LEAK TEST PROCEDURES

HTO in Collection Chamber (dis/min)	HT in Collection Chamber (dis/min)	HTO Recovered in Bubbler #1 (dis/min)	HT Recovered in Bubbler #2 (dis/min)	Tritium Recovered (%)
6.56×10^6	----	6.34×10^6	0.13×10^6	98.6
----	3.21×10^5	0.01×10^5	3.08×10^5	96.3
----	8.94×10^5	----	8.96×10^5	100.2
----	6.58×10^5	----	6.51×10^5	98.9
			Average percent recovered	98.5

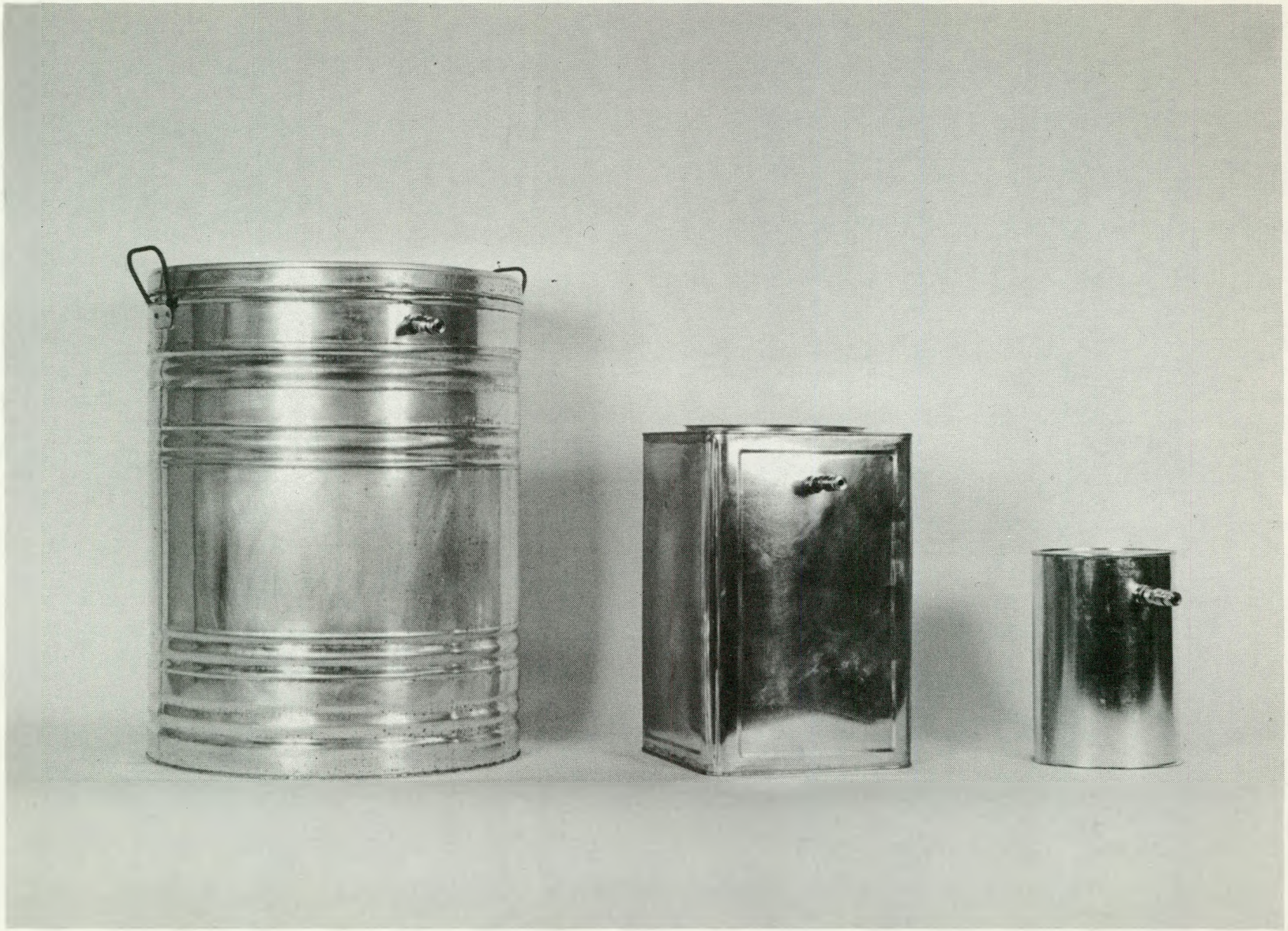

FIGURE 1 - Disposable collection chambers.

LEAK DETECTION SYSTEM

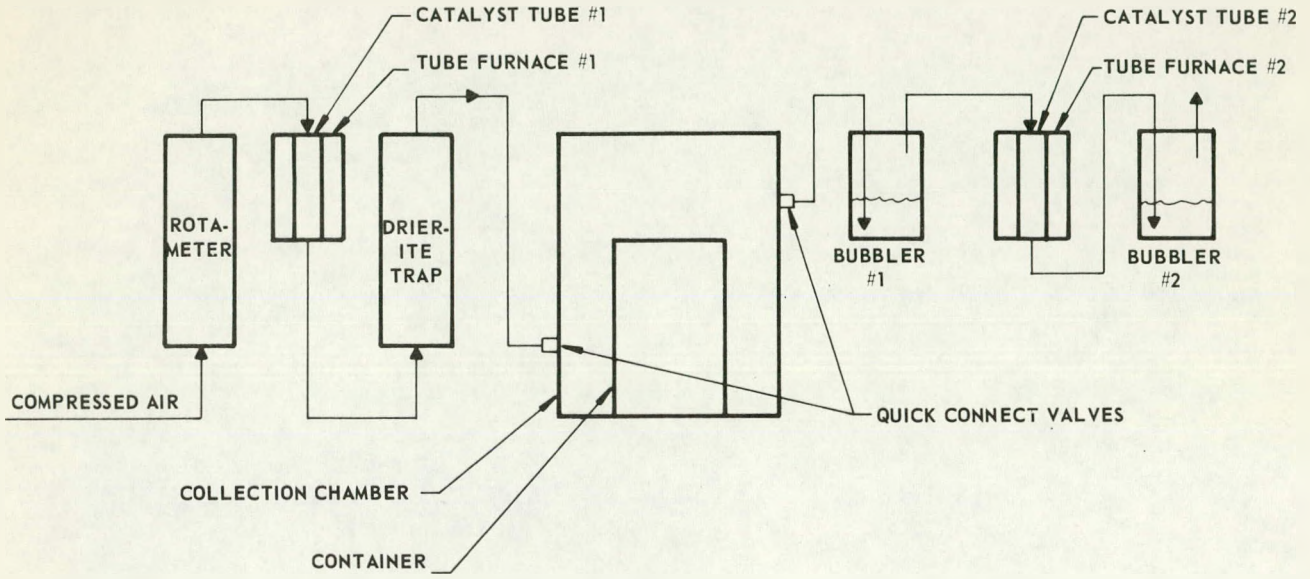

FIGURE 2

ETHYLENE GLYCOL BUBBLER

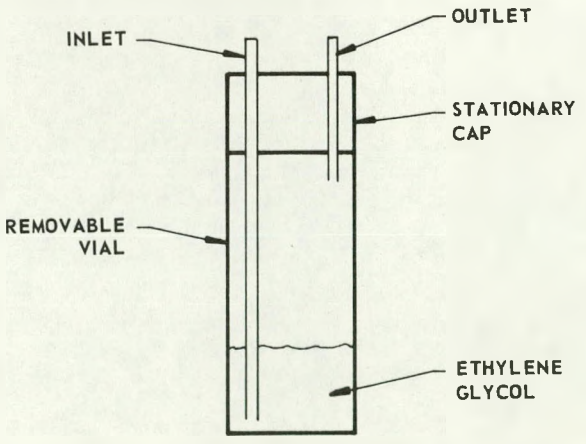

FIGURE 3