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SOME RECENT CHANGES IN TRITIUM HANDLING AND CONTROL AT MOUND LABORATORY

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SOME RECENT CHANGES IN TRITIUM HANDLING AND CONTROL AT MOUND LABORATORY

Significant reductions in tritium effluents and personnel exposures at Mound Laboratory have been made during the past 5 yr. Yearly effluents are less than 3% of former levels and personnel exposures have been reduced by a factor of 300. Several recent changes which have contributed to these reductions include lowered tritium levels in gloveboxes, and the efficiency and capacity of Mound's new effluent removal system. Personnel exposures have been reduced dramatically by changing to precious metal catalytic converters or oxidizers for use with the glovebox gas purification system. Unlike some former systems using hot copper or proprietary reactants for oxygen removal, a catalyst provides very effective removal of both oxygen and tritium. Both oxygen and tritium can be monitored and, if necessary, increments of hydrogen in argon can be added until the oxygen level is brought down to the desired value.

Effluent Removal System

Mound Laboratory's effluent removal system (ERS), a detritiation system for all types of tritium-contaminated gases, was primarily installed to receive gases from vacuum pump exhausts, passboxes, and process lines. In the last 2 yr the ERS has aided effluent reduction significantly in the areas of maintenance and decontamination. Because of its large gas collection and processing capacity, it is possible to "bag" highly contaminated lines and equipment in plastic sheeting and/or isolate them from the system by means of valves so that the equipment can be decontaminated by flushing with moist air. Maintenance is then performed in a walk-in plastic tent built around the repair area. Air in the tented area is discharged to the ERS at flow rates up to 60 ft^3/min.

Some conventional fume hoods, because of the type of work and possible loss of tritium to the atmosphere, have been modified so that the worker may have the option of venting the hood to the stack or to the ERS. Air flows must be limited to about 60 ft^3/min; hence, the opening to the hood is closed or carefully controlled to maintain proper pressure differential or air velocity across the opening of the hood.

Major problems of electrical terminal burnout and inability to sustain desired gas temperatures have been experienced with preheaters which precede the oxidizer section of the ERS. The array of internal Calrod heaters in the preheaters has been abandoned for conventional exterior band heaters which are now providing the desired 400°C gas temperatures at the exit of the preheaters.

Currently, both Hopcalite (copper oxide and manganese dioxide reactant) and platinum catalytic beds are being used in the ERS for oxidation of HT and tritiated hydrocarbons, e.g., pump oil vapors. A low temperature (40°C) filter section at the head-end of the ERS and Linde 13X molecular sieve beds downstream of the filter system were designed into the ERS to remove all types of contaminants. Since the removal efficiency to be expected at time of installation was uncertain, Hopcalite beds were installed. Hopcalite is generally less subject to poisoning than are precious metal catalysts. After several years of experience with this filter system, the removal of contaminants appears to be excellent. Therefore, heated catalytic beds have been added downstream of the four Hopcalite beds in the Mound Laboratory ERS.

These catalytic beds have given exceptional performance for both tritium and tritiated hydrocarbon removal. Even with moderate amounts of contaminants, catalytic beds would now be used in lieu of Hopcalite beds. Precious metal catalysts are not only more effective but the only readily available method for oxidizing methane. Experience with the catalytic beds shows that temperatures of 575 to 600°C are necessary to achieve total breakdown of the vapor contaminants. Condensables and particulate matter are trapped in the low-temperature filter area and trace contaminants are removed on the 13X molecular sieves. Therefore, the contaminant entering the oxidizing beds appears to be tritiated methane. Experience has shown that traces of tritiated hydrocarbons will be formed anytime when gaseous tritium comes in contact with grease, oil, or polymers.

A system to monitor the gaseous waste entering the ERS has been installed recently. The main purpose for monitoring gas flow and tritium concentrations in the waste gas is to use the information as a diagnostic tool to alert laboratory personnel of a problem which may require corrective action. In addition, if the total gas entering the ERS or its emergency storage tank should exceed its collection and processing capacity, the gas entering the system which has the lower concentration can be diverted to the stack, allowing the higher-concentration gas to be processed. The system consists of a 30-ft, 4-in. diameter header or manifold which provides the feed to the ERS and also the termination point for eight 2-in. feeder lines leading to operating laboratories throughout the building. Each feeder line is monitored with a flowmeter. For tritium assay each feeder line is sampled consecutively for a predetermined period by a programmed automatic gas sampler. Each gas sample enters a dryer for water and oil removal before entering the ionization-type monitor; this is necessary to prevent fouling of the monitor. Hence only HT concentrations can be measured.

Decontamination

The removal of tritium-contaminated equipment or containers from gloveboxes has always required special decontamination procedures. The contamination may be either dirt or tritium-containing materials. In the past, items were physically decontaminated by scrubbing and wiping with water or alcohol, which can lead to personnel exposures. More sophisticated methods using steam, heat and vacuum for decontamination are described in Tritium Control Technology, WASH-1269. Economical, simple facilities such as a vacuum passbox can serve the purpose of decontamination. In principle, the box containing the item is evacuated and about 15-25 ml of water is admitted slowly to the evacuated box through a valve so that the water is atomized as it is sucked into the box. After a minimum waiting period of 30 min, the box is placed under vacuum until dry. Using this simple method, containers held in highly contaminated gloveboxes for periods up to 5 yr have been successfully decontaminated, allowing them to be taken out into the room.

Monitoring

One of the major monitoring innovations made during the 1975-1976 period was the independent determination of HTO and HT for all stack effluents. The technique of measurement involved the collection of HTO on an absorbent or in a liquid followed by liquid scintillation counting of the collected sample.

The need for HTO and HT independent assay and the use of collection-type monitors is manyfold; 1) the biological hazard for HTO is much greater than for HT; 2) the HT/HTO ratio is considered as necessary laboratory operational information (state of the laboratory); 3) confirmation of data from real-time monitors; 4) collection monitors can provide more accurate data than do real-time monitors for routine and accidental releases; and 5) as tritium effluents are lowered to "as low as practicable" and with the large volume air flows, detection (below 5 $\mu Ci/m^3$) with real-time monitors is not possible.

One collection method is absorption on molecular sieves, followed by distillation (bake-off) and scintillation counting of the liquid. This method, which is suitable for weekly, bi-weekly or monthly collection schemes, has a detection limit of 10^{-6} $\mu Ci/m^3$.

Another low-cost versatile collection method involves the use of ethylene glycol instead of an absorbent or water (bubbler). The basic system, a version of which will be commercially available, is shown in Figure 1. Air samples are brought into the system through a filter in order to protect the flowmeter and gas totalizing meter from particulate matter. The gas totalizer is a conventional dry gas test meter with liter indexing. Air or gas samples are first passed through a series of three ethylene glycol bubblers to remove HTO (and H_2O) and then through a heated (600°C) palladium catalyst where the HT gas is oxidized to HTO and collected in a second series of ethylene glycol bubblers. Air pulled through the bubbler collection system by a metal bellows pump is discharged to the stack. Air flows are adjusted to 125-150 cc/min.

Assay of the ethylene glycol for collected tritium oxide is simple. P-dioxane liquid scintillation counting solution is added directly to the vial of glycol solution and the vials are counted directly using standard procedures.

The overall performance of any bubbler system is dependent upon its retention capabilities since collection efficiency for HTO entering the system is nearly 100% (high dilution effect). Unlike water bubblers, the glycol bubblers have a twofold advantage: 1) there is virtually no loss of HTO by evaporation and no carryover if the water-holding capacity of the glycol is not exceeded, and 2) the sample collected is in a form and a container which can be analyzed directly without further sample preparation.

For stack gas analyses, the scintillation counting vials A and D (the lead vials for HTO and HT collection, Figure 1) should contain 5-6 cm^3 of ethylene glycol. Back-up bubblers contain 10 to 15 cm^3 of glycol. Depending on the operational condition and/or requirements for data, several options are open. Samples A and D can be counted on a daily basis and saved. The remaining bubblers, which are changed weekly, can be held (not counted) for a period of 4 to 5 weeks. To obtain the total activity over a 4-week period, bubblers A, B and C (HTO) are pooled and made up to a standard volume with scintillation counting solution. Several aliquots are taken for assay. The same process is repeated for bubblers D, E and F for the HT determination. In this manner the number of assays that are required to obtain the total activity and the ratio of HTO/HT + HTO is greatly reduced while daily values are available as a guide to the effectiveness of the laboratory's effluent control efforts.

When using a 5-cm^3 ethylene glycol bubbler, 95% of the collected HTO is retained during an 8-hr period with sampling gas having a 50-60% relative humidity. The loading of the first bubbler with water (H_2O) is dependent upon the relative humidity of the sampled air. Hence, to obtain reasonable accuracy (at least 80% retention), the lead bubbler should be changed every 24 hr. With back-up bubblers B and C for HTO and E and F for HT, 99% collection and retention for 7 days of sampling can be obtained.

With a tritium detection limit of about 10^{-5} $\mu Ci/m^3$ or up to 100,000-fold lower than real-time monitors, there are many options in the use of the ethylene glycol bubbler. In terms of air concentration guide levels, sensitivity for HTO and HT of 0.01% of guide levels can easily be attained.

Glycol collection monitors have many advantages over real-time and other type collection monitors because of their versatility of application. Typical areas include: low-leak-rate determinations in which leak rates as low as 1 x 10^{-17} cm^3/sec have been measured; environmental monitoring because of the speed and high-sensitivity capabilities; stack monitoring for both HT and HTO at trace levels as well as large or accidental releases; room air monitoring which permits evaluation of laboratory tritium levels of an order of magnitude below current air concentration guide levels; and research and development applications in which confirmation of calibration gas or assays of gas samples can be easily made on a continuing basis or with batch grab samples.

Gas Transfer Pumps

Gas transfer of either waste gas or tritium gas recovered from processed waste has always been a problem because of the variable conditions of transfer and the need to assure absolute containment.

A vacuum and/or transfer pump has been under development for several years at Mound Laboratory and initial use of several units has been promising enough to warrant construction of an additional 10 units. The pump has the following characteristics: 1) vacuum of 10 µm at free air discharge; 2) vacuum of less than 50 µm at 50-psig discharge; 3) as a transfer pump, inlet pressures from 40 psig to low micron range at a discharge of 50 psig; 4) transfer capacity of 4 std ft^3/min with atmospheric pressure at intake; 5) helium leak-tightness; 6) applicability to pumping tritium because the pump oil (4-ring polyphenyl ether) undergoes low degradation and low tritium exchange; and 7) reliability expectancy of 3-5 yr without major maintenance.

The pump uses a 3/4-hp 600-rpm direct-drive motor for nonstall startup, the internal mechanism of a Welch 1402 vacuum pump, a stainless steel minimum volume housing, a heat exchanger front plate to which the pump is mounted, special shaft seals with forced lubrication, polyphenyl ether pump oil, and totally enclosed inlet and outlet filter assemblies which substantially remove all oil from the gas transfer stream. Residual traces of oil require an additional filter such as a cold trap, molecular sieves, or charcoal. Drawings and a report of the fabrication procedures and modifications are now available. The cost of the unit is about $3500 to $4000, depending upon the quantity to be produced.

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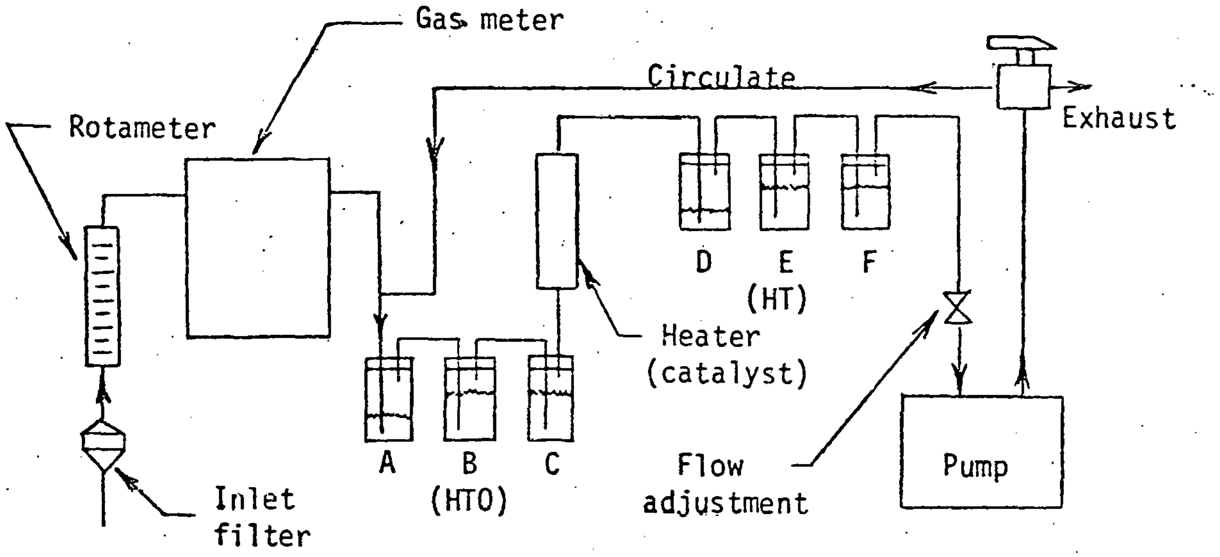

Figure 1 - Differentiating tritium collection monitor.