MONSANTO/MOUND LABORATORY ENGINEERING DEVELOPMENT OF TRITIUM-HANDLING SYSTEMS

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ABSTRACT

Mound Laboratory (Mound) has, during the past four years, been actively involved in the development of methods to contain and control tritium during its processing and to recover it from waste streams. Initial bench-scale research was directed mainly toward removal of tritium from gaseous effluent streams and from laboratory liquid wastes. The gaseous effluent investigation has progressed through the developmental stage and has been implemented in routine operations. A test laboratory embodying many of the results of the research phase has been designed and construction has been completed.

As the program at Mound has progressed, the scope of the effort has been expanded to include research concerned with handling not only gaseous tritium but also tritiated liquids. A program is presently under way to investigate the detritiation of aqueous wastes encountered in the fuel cycle of the commercial power reactor industry.

INTRODUCTION

The engineering developments described here are an outgrowth of the overall Tritium Effluent Control Project, which was initiated in 1972 when it became apparent that current state-of-the-art technology for control of tritium effluents would not result in reduction of these effluents to the RCG values at the point of emission.[1] This was the suggested guideline issued by the Atomic Energy Commission in December 1970, when contractors were requested to limit their tritium and other radioactive effluents to levels that are "as low as practicable." Between 1972 and 1976, facility design and operating philosophies were changed from dilution and release to containment and recovery with emphasis on confining the tritium at its source through the use of glove boxes and various glove box atmosphere detritiation systems. The result was a 97% reduction in tritium effluents.

Separation and enrichment technology is being developed for detritiation of both gaseous and liquid effluents. A test laboratory was designed and now is essentially completed. The laboratory consists of one main boxline (Fig. 1), which houses the developmental program equipment and the support equipment as described below.

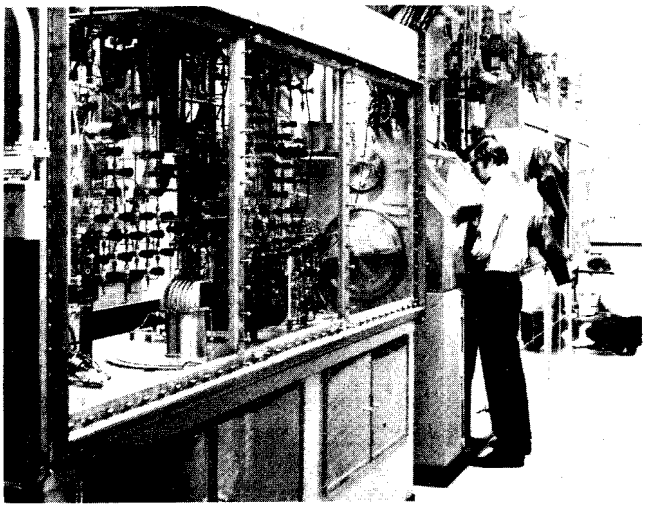

Fig. 1. Glove box line in the Effluent Control Laboratory.

GLOVEBOX ATMOSPHERE DETRITIATION SYSTEM

The Glovebox Atmosphere Detritiation System (GADS) consists of a recirculating helium purification unit that removes trace amounts of tritium along with other impurities (N_2, O_2, and H_2O) from the helium atmosphere of the glove box line. The unit operates on the principle of adsorption at liquid nitrogen temperature on a molecular sieve bed packed with Linde 5A molecular sieve. This particular adsorbent was selected on the basis of bench scale tests. A 100 ft^3/min (2.83 m^3/min) Miehle-Dexter blower circulates the helium through the purification system and back into the glove box line. Dual adsorption beds, each in their respective cold box, cycle in and out of service on a 24-h onstream and a 24-h regeneration and precool program, all automatically controlled. The entire unit is mounted on skids as shown in Fig. 2. The controls and instrumentation package, shown in Fig. 3, are located in an adjoining room. This unit has been completely cold tested and debugged. After current maintenance work is completed on the blower unit, it will be placed into service on the boxline. A unique feature of the unit is its fully automated operation requiring little operator attention.

AIR DETRITIATION SYSTEM

A second support item in the Tritium Effluent Control Laboratory is an Air Detritiation System (ADS) patterned after a larger unit that services an entire building complex at Mound Laboratory (Mound).[2] The ADS accepts effluents from the glove box line, pass box operations, and over-

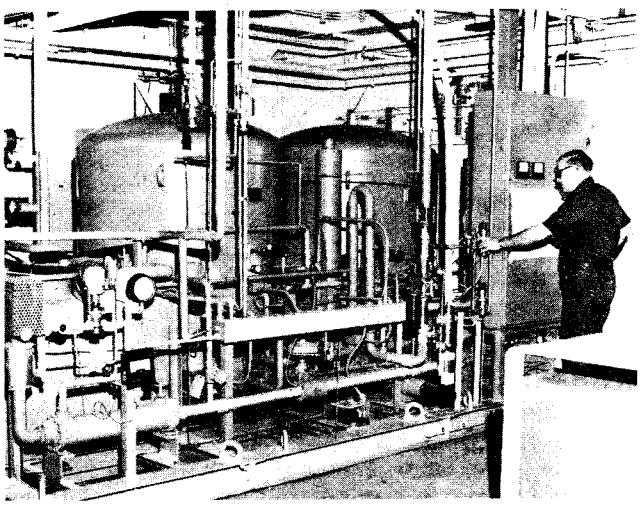

Fig. 2. The Glovebox Atmosphere Detritiation System (GADS).

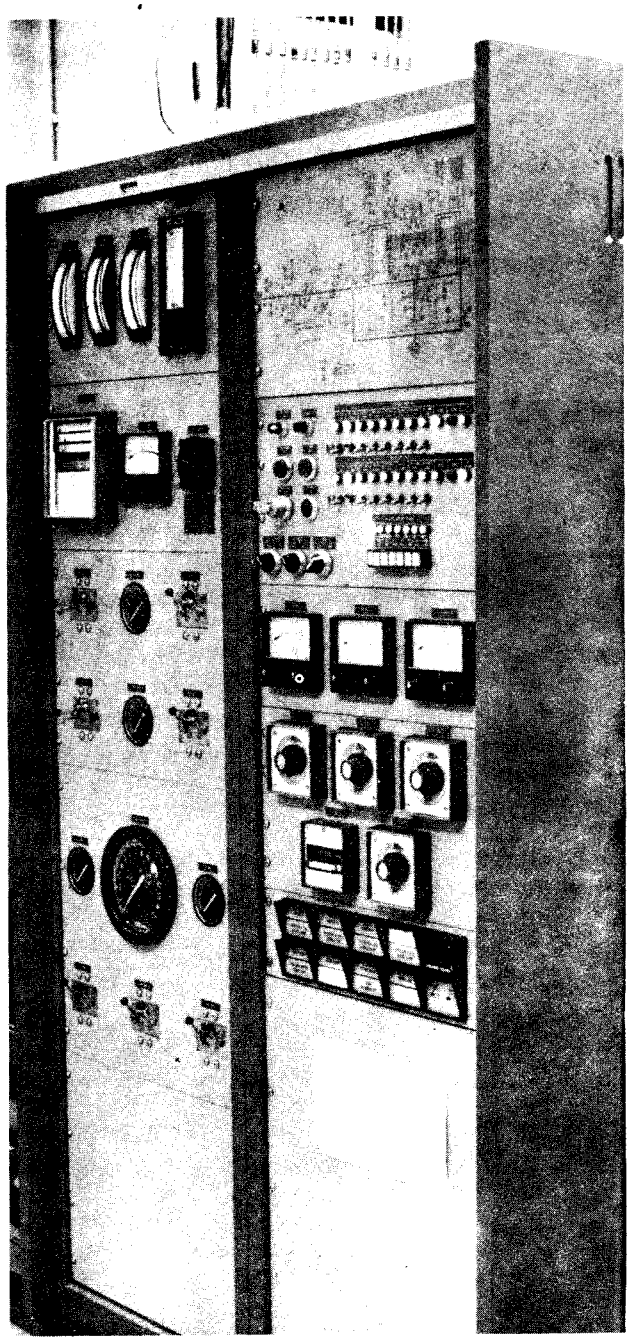

Fig. 3. The control console for GADS.

pressure vents from the GADS described above. The ADS consists basically of a platinum catalyst bed that converts the tritium in the effluent to an oxide, a drier that removes the oxide from the gas stream, and a blower that moves the gas through the system. There are two driers to permit continuous operation. The unit is capable of a 15 ft^3/min (0.42 m^3/min) throughput and is piped so that

the gas can be recirculated until the tritium level is low enough to permit stack discharge. Several tees into the inlet side of the ADS provide a hookup for ventilation of tents and other temporary enclosures used during maintenance operations. The ADS is shown in Fig. 4.

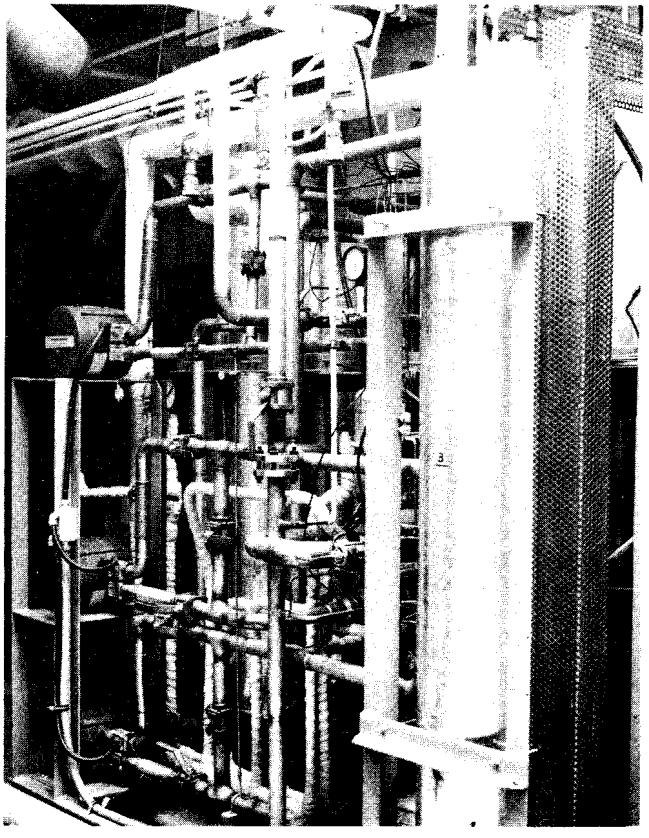

Fig. 4. The Air Detritiation System (ADS).

EMERGENCY CONTAINMENT SYSTEM

A third significant support system that was fabricated to ensure safe operation of the Tritium Effluent Control Laboratory is the Emergency Containment System (ECS).

In the unlikely event that both the primary containment within the boxline fails and the secondary containment (the boxline itself) breaches, the result could be a major release of tritium into the laboratory. This release would be detected quickly and an alarm would sound to cause evacuation of personnel from the laboratory. At the same time, dampers would close in the room ventilation system and the ECS would be activated. In its operational mode, the entire atmosphere of the laboratory is recirculated through a large platinum catalyst reactor (Fig. 5) followed by an adsorber. With the laboratory essentially sealed off, the room atmosphere is continuously passed through the ECS until the tritium level of the air permits a return to normal ventilation.

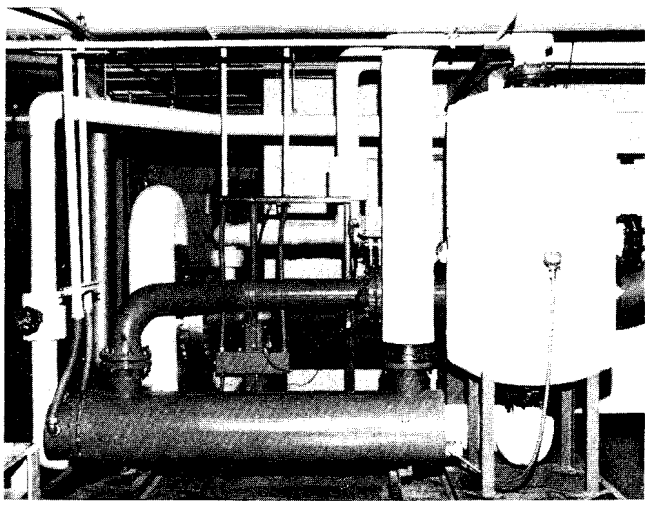

Fig. 5. A portion of the Emergency Containment System.

The 1000 scfm (28.3 m^3/min) capacity catalytic oxidizer with electric preheater and water aftercooler is combined with an adsorber section consisting of two stainless-steel vessels, each containing 3800 lb (1724 kg) of Alcoa alumina H151 adsorbent, saturated at ambient humidity. These vessels provide 10 h of operation with an inlet activity of 1 Ci/m^3 before a defined breakthrough of 100 μCi/m^3 occurs. A Spencer turbine controls gas flow.

MONITORING SYSTEMS

Three types of tritium monitoring systems are used:

1. high sensitivity stack monitors to provide the necessary information for reporting the quantity of tritium released to the atmosphere

2. room monitors to ensure personnel protection and compliance with occupational exposure guidelines

3. monitors to provide information for system control and experimental data.

All the tritium monitors use ionization chambers with vibrating reed electrometers, with the exception of a high sensitivity stack monitor that uses a gas proportional counter. Another stack monitor, parallel to the gas proportional counter, uses two 20-litre ionization chambers with a sensitivity of 0.1 μCi/m^3.

The ion chamber monitors were developed at

Mound in cooperation with an instrument manufacturer and are commercially available. A digital display reading directly in $\mu Ci/m^3$ and a logarithmic recording are used to give both a clear, prompt indication and also a permanent record. The 20-litre chambers have a sensitivity of 1 $\mu Ci/m^3$ and a range of 1 to 20,000 $\mu Ci/m^3$.

A two-level alarm system is used, which activates a flashing yellow light when the lower limit is exceeded. A flashing red light and an audible alarm are activated when the higher level is exceeded. Both of the alarms can be set anywhere between 0 and 2000 $\mu Ci/m^3$. The alarm can be silenced locally, but the lights continue to flash until the tritium concentration falls below the respective alarm points and the alarm is reset.

Electrical compensation is provided for tritium contamination of the ion chamber equal to 100 $\mu Ci/m^3$ or a gamma background of 1 mR/h. The background or amount of tritium contamination of the ion chamber can be determined by passing clean air, e.g., from a compressed air line, through the chamber instead of the normal air sample. After a steady reading is obtained, the background compensation is adjusted so that the digital display reads zero.

Information on the tritium concentration in the room air is made available to laboratory personnel by remote display units (RDU) located in the work areas. These units, which are "slaves" to the main electronic units, contain a digital display of the tritium concentration, flashing yellow and red lights, and an audible high-level alarm.

Similar monitors with 2-litre or 20-cm^3 ionization chambers are used to provide information for system controls and to provide experimental data. The range of the 2-litre units is 10 to 20,000,000 $\mu Ci/m^3$ in two steps, and the range of the 20-cm^3 unit is 0.1 to 200,000 Ci/m^3 in four steps. The major features of display, alarms, remote display, etc., are the same as on the 20-litre units.

The 2-litre chamber units are used to monitor tritium concentrations in the helium circulation system serving the glove box line, in the passbox purifier system (air detritiation system), in the exhaust duct to activate the emergency containment system, and in the cryoseparation system. A 20-cm^3 chamber used to monitor the cryogenic still system is located in the glove box line because of the high concentrations of tritium being measured.

WATER DETRITIATION DEVELOPMENT

The initial thrust of the work associated with the Tritium Effluent Control Laboratory dealt with the development of gaseous effluent control systems. Major efforts now, however, center around detritiation and recovery operations that can be applied to tritiated *liquid* waste streams. It became increasingly apparent that techniques should be developed for the removal and recovery of tritium from waste liquids that, although a relatively small volume at the present time, will show a steady rise in the next two decades. Primary sources of high-level tritiated liquid waste are the U.S. Energy Research and Development Administration (ERDA) contractors who, like Mound, have installed gaseous effluent control systems within their operations to reduce the amount of tritium being released, while at the same time generating volumes of high-level tritiated liquid waste. However, a major source of liquid tritiated waste that has yet to go onstream is the nuclear fuel reprocessing plant. These plants, which will process spent fuel from light-water reactor operations in the U.S., will be a primary source of intermediate-level tritiated liquid waste (0.01 to 1000 Ci/litre) in the near future. The Allied General Nuclear Services (AGNS) plant construction has been nearly completed, but the startup date is uncertain. Continued growth of the nuclear power industry dictates that this plant be placed in operation in the near future, followed by the construction of several additional fuel reprocessing plants by the year 2000. If we add to this the low-level tritiated liquid waste associated with the pressurized-water reactors themselves (in the cooling loops), plus an undetermined quantity of intermediate and low-level tritiated liquid waste from fusion research programs planned over the next two decades, we can see the need for the efforts initiated at Mound toward detritiation of liquid waste with subsequent recovery of the tritium.

Although several processes related to liquid detritiation (including cryogenic distillation, laser isotope separation, and electrolysis) are in various stages of development at Mound, this effort focuses on catalytic exchange detritiation. The HT/H_2O catalytic exchange process was selected because of its high separation factor and its high likelihood for success. Objectives for the catalytic exchange program have been defined as:

1. determination of the technical and economic factors of HT/H_2O exchange as a detritiation process for treating tritium-contaminated water
2. determination of the suitability of hydrophobic exchange catalysts for use in water detritiation systems
3. establishment of design criteria for detritiation systems capable of meeting ERDA

and industry needs for tritiated liquid waste treatment with particular emphasis on applications to nuclear fuel reprocessing plants and power reactors.

A preliminary cost/benefit analysis on the application of the catalytic exchange detritiation process to the 5 MT/day AGNS plant showed that the process appeared economically favorable using the guidance of $1000 per total body manrem for the benefit figure.

During the past $1\frac{1}{2}$ years, Mound and Engelhard Minerals and Chemicals Corporation have worked together to design and fabricate (at Engelhard) an experimental apparatus suitable for glove box installation in the Tritium Effluent Control Laboratory (Fig. 6). The unit has been installed, leak tested, and checked out operationally with cold test programs. Corrective steps were taken to make the system function as designed. Analytical instrumentation has been integrated into the system as seen in Fig. 7.

This apparatus is to be used for catalyst evaluation tests. The first catalyst to be evaluated will be an Engelhard hydrophobic catalyst consisting of platinum on an alumina substrate with a semipermeable coating of a proprietary water repellent material. This type of catalyst permits direct contact exchange reactions between liquid water and gaseous hydrogen at room temperature where separation factors for this reaction are quite favorable, Isotope Separation Factor (ISF) = 6 at 25°C. The experimental unit consists of

1. a low-temperature reactor (20 to 70°C)
2. a high-temperature reactor (100 to 500°C)
3. an evaporator
4. a condenser
5. a phase separator
6. pumps
7. analytical instruments.

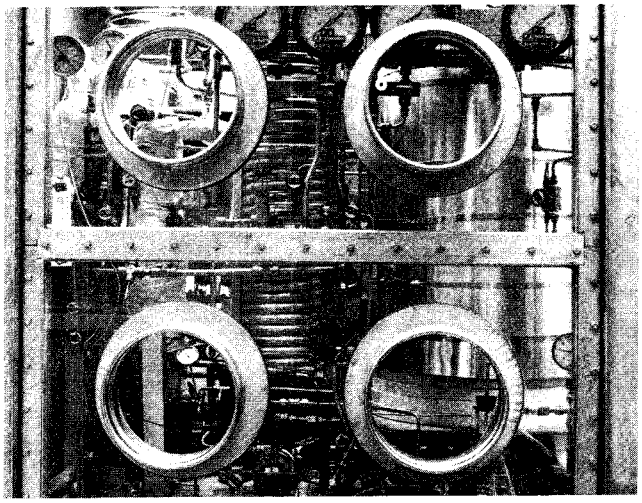

Fig. 6. Experimental setup for catalyst evaluation.

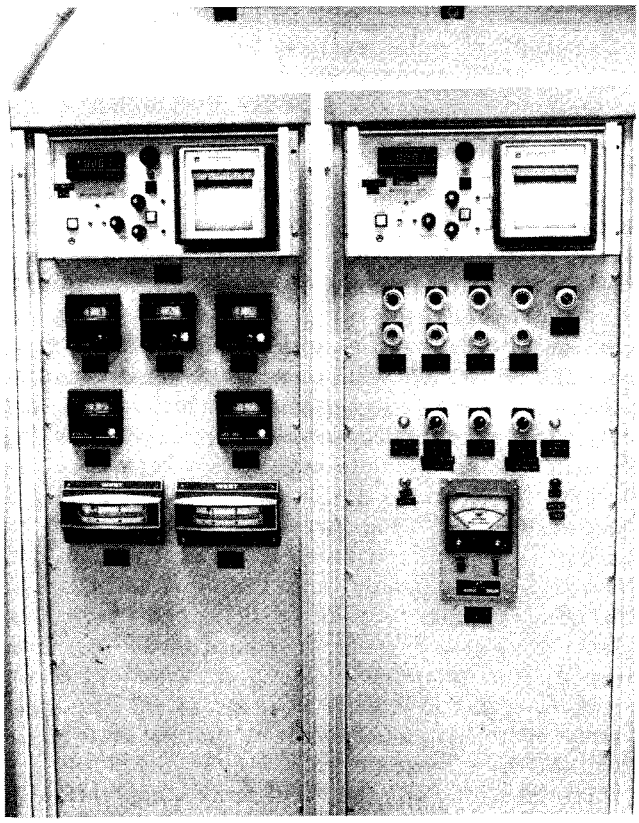

Fig. 7. Instrumentation for catalyst evaluation.

The low-temperature reactor contains the hydrophobic catalyst and in it tritium is enriched in the water stream and depleted in the gas stream in countercurrent flow. The high-temperature reactor contains a nonhydrophobic platinum catalyst, and tritium is depleted from the water vapor in a co-current H_2O/HT vapor system. The system has a completely closed loop for study of gas phase-liquid phase tritium exchange reactions. Flow rates and temperatures may be varied and reaction rates may be measured under steady conditions. Initial tests will be conducted with tritium concentrations of 0.01 to 1.0 $\mu Ci/cm^3$. Kinetic and thermodynamic performance data will be obtained and evaluated not only on the Engelhard catalyst, but also on a catalyst developed by Atomic Energy of Canada, Ltd. (AECL), who have

an interest in the catalytic exchange detritiation process for their heavy water reactor program. In 1975, ERDA and AECL began negotiations for a cooperative program to share information concerning the catalytic exchange detritiation process.

CONCLUSION

The tritium-handling systems described will be operated to gain experience and data relevant to defining the "as low as practicable" criterion.

If the HT/H_2O hydrophobic exchange catalyst evaluation tests are successful, a multistage pilot unit will be designed and constructed to further evaluate process parameters and controls.

ACKNOWLEDGMENT

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