

TRITIUM SYSTEMS TEST ASSEMBLY (TSTA) STABILIZATION

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The Los Alamos National Laboratory (LANL) Tritium Systems Test Assembly (TSTA) project was begun in 1978 to develop, design, and demonstrate the technology and safe operation of selected tritium processing systems required for a fusion reactor. In 2001, the US Department of Energy (DOE) determined that TSTA's mission was complete and that the facility should be stabilized.

At the completion of the stabilization project in 2003, TSTA was categorized as a radiological facility. Before stabilization was complete, the tritium inventory at TSTA was grouped in the following categories: tritium gas mixed with hydrogen isotopes, tritiated water absorbed on molecular sieve, tritium held up as a hydride on various metals, and tritium held up in process components. For each of these, tritium content was characterized, a path for removal was determined, and the proper disposal package was developed. Hydrogen exchange, calorimetry, direct sampling, pressure/composition/temperature, radiological smear surveys, and controlled regeneration were used to determine the tritium inventory for each category of tritium.

After removal, the tritium inventory was either (1) sent to other facilities for reuse processing or (2) buried at the LANL radioactive waste disposal site. One complete experimental system was packaged and transferred to another DOE site for future use. Special burial containers were designed and fabricated for the inventory buried at the LANL radioactive waste disposal site. The project was conducted with low tritium emission to the environment and negligible personnel exposure. After the tritium removal was complete, all remaining hardware and piping were opened and vented; the facility emission was below 1 Ci per day.

I. INTRODUCTION

The TSTA project was begun in 1978 to develop, design, and demonstrate the technology for and the safe operation of selected tritium processing systems required for a fusion reactor. The TSTA is located at LANL in Los Alamos, New Mexico, and initially was funded by the US DOE. Tritium processing began at TSTA in 1984.

In 2001, DOE determined that TSTA's mission had been completed successfully and the facility should be stabilized. Stabilization involved placing TSTA in a safe, stable configuration with a goal of reducing the tritium inventory to below the DOE low-hazard nuclear facility threshold of 16,000 Ci. The facility was to be maintained in this state until funding was available for the final decontamination and decommissioning (D&D). This paper describes the process used and the results of the activities required to achieve the safe, stable condition.

At the completion of the TSTA operation mission, the tritium inventory at TSTA was 140 grams. The facility was categorized as a DOE moderate-hazard nuclear (Category 2) facility

II. CHARACTERIZATIONS

High-surface-area components (molecular sieve, uranium, etc.) were characterized using one of the methods described below.

II.A. Hydrogen Exchange

Uranium beds or other heated metal beds can be measured using a technique known as hydrogen exchange. This technique commonly is used following a standard bake off (450°C for several hours). A heel of X moles of hydrogen isotopes is left on the uranium bed with an unknown tritium isotopic concentration. To assess the quantity of tritium in the heel, a quantity of protium (which depends on the size of the bed) is first sorbed onto each metal bed; alternately, a sample of the original gas desorbed from the bed can be taken when the bed is heated. If protium is added, the beds are heated to 350°C to promote the exchange between the protium that was added and the tritium on the heel. Each bed is baked off to 450°C, and a sample of the gas is collected. The gas is analyzed for tritium content using a mass spectrometer or BSD. At the bakeoff conditions, the gas collected should have the same tritium fraction as the gas that remains in the uranium (heel) (TI mole %).

A known quantity of protium (Y moles) then is added to each heated metal bed. The bed is heated to 350°C again to promote equilibrium between the protium that was added and the remaining heel. The bed then is desorbed to the standard bakeoff temperature, 450°C, and

a second sample was collected. The quantity of hydrogen desorbed should be Y moles, leaving the quantity of hydrogen in the heel the same. The second sample is analyzed for tritium ($T2$ mole %) and is assumed to be representative of the tritium concentration in the diluted heel. The quantity of tritium before and after the exchange can be described as

$$X * T1 = (X * T2) + (Y * T2) .$$

Rearranging the terms, the number of moles of hydrogen in the uranium heel is:

$$X = (Y * T2) / (T1 - T2) .$$

The total tritium remaining on the heel is

$$X * T2 \quad [1]$$

II.B. Helium-3 and -4 Ratio

The component is filled with helium-4, allowed to equilibrate, and then evacuated. This process is performed 4 times to minimize the quantity of helium-3 generated by tritium decay before the measurement. The excess helium-4 should displace most or all of the helium-3 trapped in the molecular sieve. After the fourth evacuation, helium-4 is added a fifth time. The fifth addition uses a calibrated volume of helium-4 to determine the exact quantity added. After at least 30 days of tritium decay have passed, a gas sample is taken and submitted for mass spectrometry analysis to determine the relative abundance of helium-3 in proportion to helium-4. The total quantity of helium-3 generated by tritium decay is determined by multiplying the helium-3/helium-4 ratio by the total quantity of helium-4 added initially. As an illustration of the rate of helium-3 production, note that 1 mole of tritium gas (T_2) molecules (6.032 grams) would generate 0.01075 moles of helium-3 atoms in 35 days. The quantity of T_2 in the component can be calculated based on the amount of helium-3 generated over a specific time frame [2].

III.C. Direct Sampling

The dry bulk material (molecular sieve, vermiculite, etc.) is sampled, its volume and weight are determined, and a known quantity of water is added to the sample. After a minimum of 24 hours, a pipette sample (usually 10 μ l although it can be as much as 1 ml) of the water is taken, 10 ml of Ultima gold solution are added, and the sample is analyzed in a Packard liquid scintillation analyzer. Depending on the activity in the pipette sample, the original sample may have to be diluted to reduce its activity. This is accomplished by taking a 1-mL or smaller pipette sample from the original pipette sample with the

Ultima gold solution, placing it in a clean count vial, and adding 10 ml of Ultima gold. When the analyzer provides valid numbers, the dilutions are corrected back to the original pipette sample. Because the volume of the material is known, the sample activity can be calculated for the entire container. Liquid samples are counted directly without the water addition.

II.D. Controlled Regeneration

The molecular sieve bed was heated in the normal method for regeneration, the helium purge gas and water vapor were passed across a heated magnesium bed, and a sample the gas stream on the outlet of the magnesium bed was taken. The gas stream contained trace amounts of impurities, HDT, and the helium carrier gas. The amount of tritium on the molecular sieve can be determined using temperature of the molecular sieve bed and the tritium ratio. The water content of the molecular sieve can be estimated using published adsorption isotherm data under measured conditions of temperature and water vapour pressure. Water vapour pressure can be measured in two ways: (1) a dew point probe placed in the purge gas outlet can be related to pressure directly or (2) a gas sample can be taken for analysis in a mass spectrometer. Water vapour is difficult to measure accurately because of its affinity for most surfaces that it contacts, so we cracked the water vapour over heated magnesium and used the liberated hydrogens (Q) to determine both the total water vapour pressure and the isotopic concentration of tritium in our measurements [3].

II.E. Calorimetry

Heat of decay can be used to determine the total tritium activity in the component.

II.F. Pressure-Volume-Temperature

When the bulk gas was being dispositioned it was sampled and its composition was determined on a mass spectrometer or a beta scintillation detector to allow a tritium inventory to be calculated.

III. WASTE DISPOSAL METHODS

All of the waste generated at the TSTA facility was disposed of at TA-54, which is a radioactive waste repository at Los Alamos. The waste generated fit into one of the four categories listed below.

- Compactable, less than 20 μ Ci/m³, off gassing waste consists of routine daily waste that is compacted before final disposal.

- Non-compactable, less than 20 mCi/m³, waste consists of larger items that do not meet the compactable requirements and have less than 20 mCi/m³ activity.
- DOT 7A type A drums, < 500 Ci/package, are used for process waste. If a piece of piping was removed from a tritium gas handling system, it would be weighed, assigned a curie value based on weight, and disposed of in the drum. The curie-to-weight ratio is 5 Ci/kg, which was determined through analysis at the Savannah River Site (Reference 4).
- Stainless-steel overpacks, < 10 grams H₂O, are used for high-activity items that cannot be disposed of through the routine methods listed previously. Four different ASME pressure-rated vessels fabricated from stainless steel were used.

The facility tritium inventory was approximately 140 grams when the decision was made to halt operation and start the stabilization process. Table I shows the steady decrease in tritium inventory throughout the stabilization process.

Table I. Tritium Reduction.

Material Type	Quantity (grams)	Removal Date
Product gas	15	2000
Bulk gas	71	2001
Legacy moisture collectors	31	2002
Waste (02)	1	2002
Waste (03)	21	2003
Contamination	0.5	N/A
Total Quantity	140	

The tritium inventory on legacy moisture collectors was approximately 31 grams, the individual quantities in some of the moisture collectors required special welded overpacks to safely package the waste. Fig.1. shows the loading process of one of the 20 overpacks loaded during the legacy waste campaign. Fig.2. shows the final assembly of the overpack prior to welding. Fig.3. shows an overpack being lowered into the burial shaft for long-term storage. Fig.4. shows the transportainer and racks used for overpack handling and shipping.

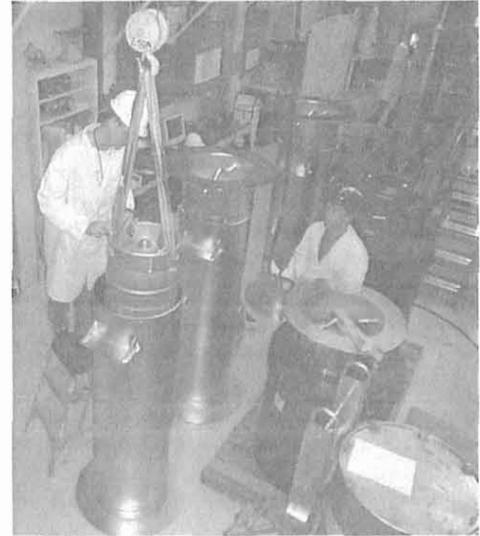


Fig.1. Loading Legacy Moisture collectors.



Fig.2. Assembling Overpacks.



Fig.3. Burial Shaft.

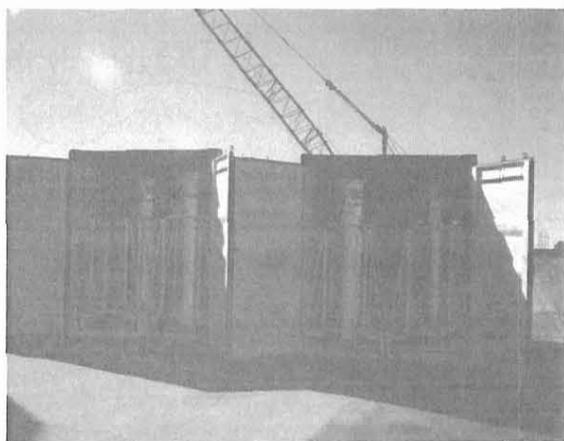


Fig.4. Overpack Shipping Containers

IV. DEPLETED URANIUM OXIDATION

One of twelve gloveboxes in the process system contained five 6 kilogram depleted uranium beds for solid storage of hydrogen isotopes with different concentrations of tritium. The bulk gas on the uranium beds was stored on a Hydride Storage Vessel (HSV) and sent to the Savannah River Site for future use. The tritium heel on the uranium beds was determined using the hydrogen exchange method. The final tritium inventory on each bed was low enough to allow disposal in a 55-gallon stainless steel drum. The beds were oxidized *In situ* using

gas with less than 5% oxygen in argon or helium. The flow rate of the oxidation gas was limited based on the bed temperature, the maximum flow rate was 5 liters per minute. The oxidation was determined to be complete when the uranium bed was at its final operating temperature with no increase in temperature from the heat of oxidation.

Six smaller beds with less than one kilogram of depleted uranium were oxidized using the same process. Calorimetry was used to determine the amount of tritium remaining on the smaller uranium beds prior to oxidation.

V. CONCLUSIONS

The stabilization project for the TSTA facility was a success for the Los Alamos National Laboratory. The project was carried out safely with minimal stack emissions and minimal personnel exposures; it was completed on schedule and under budget.

REFERENCES

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