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HYDRIDE BED ISOTOPIC EXCHANGE

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Savannah River Tritium Enterprise (SRTE) has used $LaNi_{4.25}Al_{0.75}$ (LANA75) hydride beds to store hydrogen isotopes for over 2 decades.

A benefit of using LANA75 is that the helium-3 generated from tritium decay is retained in the hydride material, allowing the hydride beds to deliver high purity product gas. A disadvantage is that the helium-3 accumulates in the LANA75 material over time, which forms a heel that cannot be removed under normal operating conditions. The heel traps hydrogen in the bed, slowly reducing the operational capacity of the bed as the heel grows. Eventually, the helium-3 begins to release from the material, preventing the delivery of high-purity product.

The hydride beds are replaced when: 1. operational capacity is reduced such that it is impactive to routine operations, and/or 2. product purity is not maintained due to helium-3 release. Prior to replacing and disposing of the beds, it is necessary to isotopically exchange the gas on the bed to recover as much tritium as possible. Isotopic exchange involves repeatedly absorbing deuterium onto the bed and desorbing hydrogen isotopes from the bed, until a pre-determined criterion has been met. The isotopic exchange process represents a significant additional load on routine operations both in time and in the amount of waste gas that requires further processing.

A set of beds was recently prepared for replacement. The isotopic exchange method used by SRTE is presented, along with results of the most recent isotopic exchange. Lessons learned during the recent isotopic exchange process led to modifications that reduce isotopic exchange duration and corresponding waste gas produced, while increasing the amount of tritium recovered.

I. INTRODUCTION

The Savannah River Tritium Enterprise (SRTE) uses metal hydride storage beds for process gas absorption, storage, and desorption [1, 2]. Over time, as tritium stored on the beds decays, its byproduct, helium-3 accumulates in the hydride material. This accumulation of the helium-3 has both advantages and disadvantages. For many years, the hydride material retains the helium-3 and therefore allows delivery of high-purity tritium. However, the helium-3 eventually accumulates to a point where it begins to release from the material. In addition, the helium-3 in-growth traps a portion of the hydrogen, which can reduce the operational reversible storage capacity of the beds to approximately 30% - 40% of their original capacity. The heel can result in a tritium hold-up of approximately 30% of the bed's capacity at the end of its service life. Details of LANA75 hydride bed aging have been reported previously [3, 4, 5].

Tritium and helium-3 are valuable resources, and it is therefore necessary to recover them prior to disposing of the beds. Since current bed design does not allow heating the bed to temperatures required to remove the entire heel from the bed, another method involving isotopically exchanging the heel to recover tritium was developed [6].

During the most recent performance of the isotopic exchange process, some obstacles were encountered that led to altering the process. This paper reviews the methodology for performing isotopic exchange, and describes changes made to improve the methodology. Improvements resulting from these changes include an increase in tritium recovery, reduction in time required to perform isotopic exchange, and a reduction in waste gas produced.

II. METHODOLOGY

Historically, the isotopic exchange process involves absorbing deuterium onto a cooled bed, then heating the bed to desorb the deuterium/tritium mixture. These absorptions and desorptions are repeated until the tritium is reduced to a pre-determined target or until no additional tritium is removed from the bed.

Figure 1 is a simple depiction of this process. A measured quantity of absorption gas (deuterium) is sent from a tank to the cooled bed where it absorbs into the bed and exchanges with a portion of the tritium. The resulting mixture of deuterium and tritium is then desorbed to a tank, where quantities and compositions are

measured. Pressures are measured using pressure transducers (e.g., strain gauge), temperatures are measured using thermocouples and resistance temperature detectors (RTDs), and gas compositions are measured using mass spectrometry. A material balance is then performed to determine the remaining quantity of hydrogen isotopes on the bed. Once a pre-determined threshold is reached, or the cost and effort to remove additional tritium exceeds the value gained, the cycles are terminated.

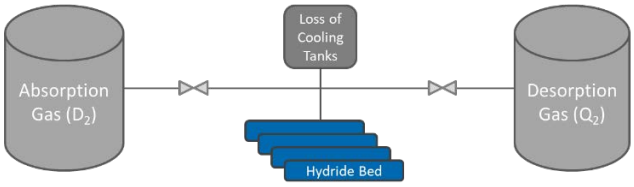

Figure 1. Depiction of isotopic exchange process.

While performing isotopic exchange to prepare for a recent bed replacement, it was observed that, after 6 exchange cycles, a significant portion of the tritium heel remained on the bed and that additional exchanges were not likely to remove additional tritium.

These data are summarized in Figure 2 below.

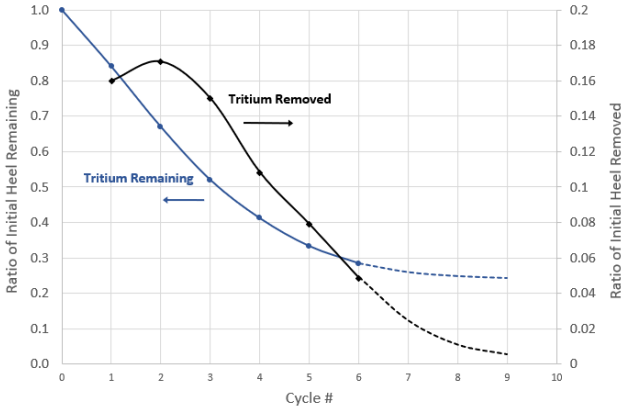

Figure 2. Initial results and projections.

In the figure, the tritium removed from the bed and tritium remaining on the bed are shown as a function of the number of exchange cycles. The vertical axes have been normalized such that a value of "1.0" corresponds with the initial quantity of tritium forming the bed's heel.

The figure shows a steady decrease in the amount of tritium removed each cycle. At the end of cycle 6 the quantity of tritium removed per cycle was reduced to approximately 4% of the initial heel amount. It is reasonable to assume that additional cycles would not have removed significant amounts of tritium and that the final tritium content would have approached a steady value around 25% of the original heel, as depicted by the dotted lines. Leaving a heel of this magnitude on the bed was unacceptable to the facility given the value of tritium and the need to prepare the bed for safe disposal.

Following collaboration between Tritium Facilities and Savannah River National Laboratories (SRNL), the decision was made to heat the bed within a confined volume following absorption of deuterium to encourage exchange between the deuterium and tritium. Accordingly, the bed was heated following deuterium absorption for Exchange Cycles 7 and 8, and notable differences from previous exchanges were obtained. Results are presented in the next section.

After performing the 8th exchange cycle, it was decided to vary the approach even further. As depicted in Figure 1, hydride beds are connected to loss-of-cooling tanks, which form an integral volume with the beds (i.e., there is no isolation between beds and loss-of-cooling tanks). Following the absorption of deuterium during Exchange Cycles 9 – 12, the bed was cycled between two temperatures repeatedly to drive gas from the material to the loss-of-cooling tanks and back to the material on the bed. The upper temperature was chosen to correspond with system pressure limitations, and the lower temperature was chosen to be 80ºC. It has been observed that above 80ºC, LANA material has some preference for the lighter isotope and that below 80ºC the LANA material prefers the heavier isotope [7]. During Exchange Cycles 9 – 12 it was desired to maintain temperature above 80ºC to take advantage of the preferential absorption of lighter isotopes over 80ºC.

IV. RESULTS & DISCUSSION

Figure 3 shows the results of heating the bed to encourage exchange of deuterium and tritium for Cycles 7 and 8.

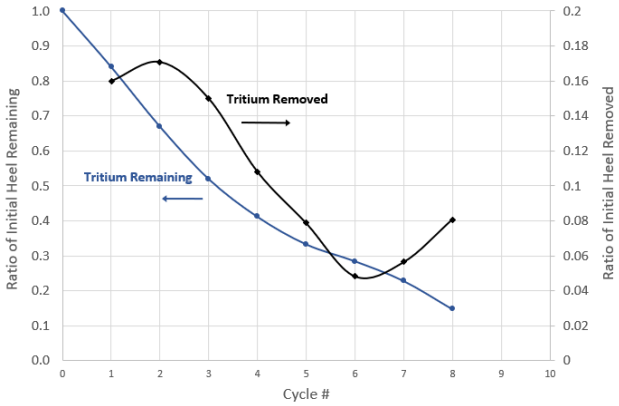

Figure 3. Results following heated exchanges.

During Cycle 7, the bed was heated for approximately 4 hours following absorption of deuterium. It is clear from the figure that heating promoted deuterium-tritium exchange in the bed, as evidenced by the increase in the amount of tritium removed from the bed relative to previous exchange cycles. During Cycle 8, the bed was heated again, but this time overnight. Again, the amount of tritium removed increased relative to previous cycles.

For the remaining cycles (Cycles 9 – 12), the absorption gas was cycled between the bed and loss-of-cooling tanks approximately 10 times by cycling between two temperatures (above 80ºC). Results are shown in Figure 4.

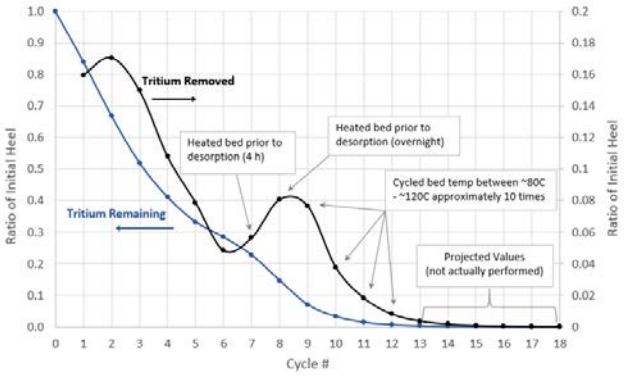

Figure 4. Results of cycling bed between 80ºC and 120ºC.

This figure shows the reduction of the heel to approximately 1% of the original heel at Cycle 12. The tritium remaining on the bed during the final 4 cycles appears to follow a theoretical reduction assuming perfect mixing of the absorption gas with the heel gas. This may be attributed to the fact that the cycling performed following absorption essentially equates to several additional cycles and that it takes place at temperatures that encourage exchange and preferential absorption of the lighter isotope in the hydride material.

VI. CONCLUSIONS

The isotopic exchange process is a critical process to perform at the end of a hydride bed's service life to recover tritium and prepare the bed for disposal. The process comes with a cost to the facility. In addition to the need for resources and personnel, each exchange generates a mixture of deuterium and tritium that requires further separation.

During a recent bed change-out, the following lessons were learned, and improvements have been made to the isotopic exchange process.

First, heating the bed following absorption of the deuterium flush gas promotes exchange between the deuterium and tritium.

Second, moving the gas between the bed and loss-of-cooling tanks by cycling bed temperature essentially results in performing several more exchange cycles more efficiently. This reduces the overall number of exchanges and the time to perform the isotopic exchange process.

Finally, maintaining the temperature above 80ºC during this cycling promotes deuterium-tritium exchange since the material favors the deuterium above this temperature.

The lessons learned and changes made to the isotopic exchange process described above resulted in recovering approximately 20% - 25% of a tritium heel that would not otherwise have been recovered during a recent bed change-out. It also reduced the number of overall exchanges and the amount of waste gas produced while significantly improving the efficiency of the exchanges.

These improvements will be incorporated into facility procedures to increase efficiency of isotopic exchange on all hydride beds in the future.

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