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Hydride Bed Helium-3 Recovery and Partial Regeneration

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Abstract — Savannah River Tritium Enterprise has used $\text{LaNi}_{4.25}\text{Al}_{0.75}$ (LANA75) hydride beds to store hydrogen isotopes for over two decades. A benefit of using LANA75 is that the ^3He 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 ^3He 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 ^3He 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 ^3He release.

Several beds were operated beyond their design life. One of these beds was selected to undergo heating beyond its normal operating temperature to evaluate the possibility of removing a portion of the hydrogen and helium heel to improve bed function until a replacement could take place. This bake-out removed a portion of the hydrogen and helium heel, and preliminary data indicate that bake-outs may partially regenerate the beds. The bed's performance will continue to be monitored, and additional bake-outs will likely be performed. Performing bake-outs results in increasing the recovery of ^3He , more efficient end-of-life activities (such as isotopic exchange), and extension of the useful service life of the bed.

Keywords — Hydride, regeneration, helium, tritium.

Note — Some figures may be in color only in the electronic version.

I. INTRODUCTION

The Savannah River Tritium Enterprise uses metal hydride storage beds for process gas absorption, storage, and desorption.^{1,2} Over time, as tritium stored on the beds decays, its by-product, ^3He , accumulates in the hydride material. This accumulation of the ^3He has both advantages and disadvantages. For many years, the hydride material retains the ^3He and therefore allows delivery of high-purity hydrogen. However, the ^3He eventually accumulates to a point where it begins to release from the material. In addition, the ^3He in-growth traps a portion of

the hydrogen, which can reduce the reversible storage capacity of the beds to approximately 30% to 40% of their original capacity. The heel can result in a tritium holdup of approximately 30% of the bed's original capacity at the end of its service life. Details of $\text{LaNi}_{4.25}\text{Al}_{0.75}$ (LANA75) hydride bed aging have been reported previously.^{3–5}

Replacing aged beds is expensive and requires long outages that require coordination with other facility outages and priorities. Complexities associated with these replacements sometimes result in a bed being used beyond its recommended service life. When this happens, it is necessary to contend with the reduction in capacity and ^3He release.

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Several beds operated well beyond their expected service life and showed signs of aging. To manage ^3He release, normal desorption temperatures were lowered to reduce the amount of ^3He released from the hydride material. Lowering desorption temperatures also reduces the amount of hydrogen removed from the bed, which further reduces usable capacity. An effort was made to remediate an aged bed by heating it beyond normal operating temperature. The goal of heating to elevated temperatures was to drive some of the ^3He heel out of the material to restore partial capacity to the bed. This paper describes the bed bake-out process and its impact on ^3He recovery and bed performance.

II. EXPERIMENTAL

The hydride bed used in the bake-out was a Forced-Atmosphere Cooling, Electrically Heated (FACE) bed. The FACE bed uses glove box atmosphere flowing through the annulus to cool the bed during absorption and electric heaters to heat the bed during desorption. The bed is insulated during desorption by evacuating the annulus. The FACE bed has been described previously⁶ and is shown in Fig. 1.

Interlocks on the heaters protect the bed from overheating and subsequent overpressurization in the event of a runaway heater scenario. To perform the bake-out, interlocks were raised to accommodate a higher temperature, still within the design temperature of the vessel and associated instruments and equipment. Thermocouples were attached to the outer vessel and to surrounding process piping to monitor temperatures. A small fan was installed near the bed to dissipate heat from surrounding piping connections to ensure temperatures did not exceed design temperatures.

The following methodology was used to perform the bake-out and evaluate its impact:

Step 1: A bed was filled to normal operational capacity.

Step 2: It was then desorbed to a reduced desorption temperature (to avoid ^3He release), and gas was sent to a tank to measure pressure, temperature, volume, and composition.

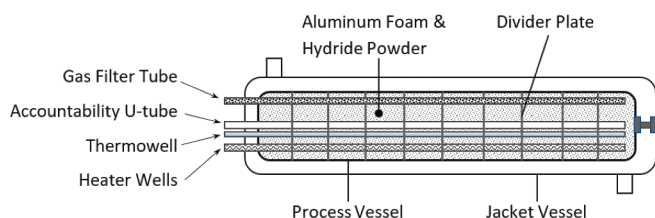


Fig. 1. FACE bed.

Step 3: In-bed accountability, a calorimetric process used to measure quantity of tritium on a hydride bed, was performed to document the amount of tritium remaining on the bed.^{7–10}

Step 4: Temperature was increased in increments to desorb gas from the bed. Gas released from the bed was pumped to a known volume for pressure, temperature, and composition measurements. In this step, bed temperature was increased above normal desorption temperatures.

Step 5: When maximum temperature was reached, the desorption was terminated, and the bed cooled. Final pressure, temperature, volume, and composition were measured in the receiving tank.

Step 6: In-bed accountability was repeated to measure the remaining tritium heel on the bed.

The entire evolution lasted approximately 2 days; however, this time may be shortened to 1 day in subsequent bake-outs.

III. RESULTS AND DISCUSSION

Figure 2 shows total gas (tritium, ^3He) desorbed from the bed along with the bed temperature during desorption. The amount of gas desorbed is normalized as a percentage of total gas desorbed. On the x-axis, the duration of zero corresponds to the beginning of step 4 of the methodology (i.e., after the initial desorption described in step 2 was complete).

As seen in Fig. 2, the bed was heated slowly over the course of 30 h to avoid potential pressure excursions from rapid release of ^3He and tritium from the material. In addition, a metal bellows pump was used to evacuate desorbed gas. These precautions resulted in a maximum pressure of 2.2 psia on the bed, well within operational

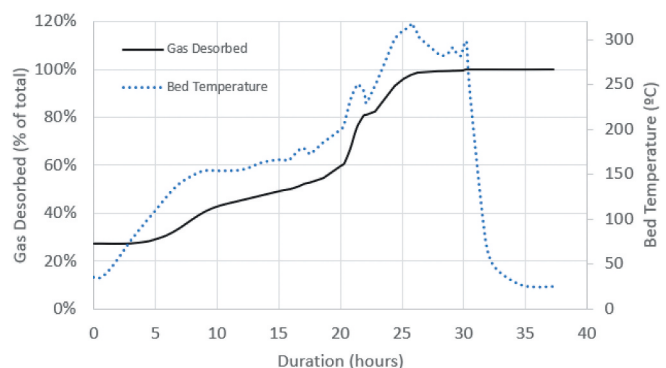


Fig. 2. Gas desorbed and bed temperature.

limits. The maximum temperature reached during the bake-out was approximately 320°C.

The line representing the amount of gas desorbed starts at 27%. This corresponds to the amount of gas removed prior to heating above the reduced desorption temperature and consisted primarily of tritium. An additional 13% of the gas removed was removed when the bed was heated to its normal desorption temperature. The final 60% of the gas desorbed was removed at elevated temperature.

A sharp rise in both the temperature and gas removed is seen starting at 20 h. This is due to an increase in heater output. For the first 15 to 20 h of the bake-out, the bed temperature was increased by adjusting the target temperature of the bed. At approximately 15 to 20 h, the temperature increase was controlled by adjusting the heater output directly. Since there was no pressure buildup in the system, the heater output was ramped up more quickly during the latter portion of the bake-out.

Gas desorbed during the bake-out was sent to a tank to monitor the quantity and composition of gas removed from the bed. It was noted previously that most of the gas removed prior to elevating the desorption temperature was tritium, as expected. The gas that was removed above the reduced desorption temperature was a mixture of tritium and ^3He . Over 60% of this gas was ^3He . This is significant since this represents ^3He and tritium recovered from the bed that would not otherwise have been recovered during routine operation.

The impacts of baking out the hydride bed were both immediate and long-term. The immediate impact was the recovery of significant quantities of both tritium and ^3He previously unavailable to the facility. The long-term impact of the bake-out is measured in two ways: monitoring reversible capacity following the bake-out and monitoring ^3He in desorbed gas.

The reversible capacity is monitored by comparing the amount of gas desorbed from the bed following the bake-out to the amount of gas desorbed prior to the bake-out. Preliminary data indicate that the reversible capacity of the bed following the bake-out increased by an average 61% relative to the reversible capacity immediately prior to the bake-out. Although reversible capacity was not restored to original bed capacity, an average 61% increase in capacity is a significant improvement.

The amount of ^3He in the desorbed gas is another way to monitor the long-term impact of the bake-out. Prior to the bake-out, the desorption temperature was reduced to prevent ^3He release from the material. Following the bake-out, the bed was heated to normal

desorption temperatures, and the amount of ^3He release has been well within the required thresholds. It is expected that the significant improvement in performance will decrease over time as the bed continues to be exposed to tritium and ^3He accumulates in the material. However, additional bake-outs could potentially restore bed performance.

Based on the results thus far, the hydride bed was partially regenerated. It is not considered a full regeneration since temperatures could not be reached that would drive the remaining ^3He heel from the material.¹¹

IV. CONCLUSIONS

It was shown that a FACE bed can be heated above normal operating temperatures to partially regenerate the bed. This bake-out of the bed removes a portion of the ^3He heel and restores some capacity to the bed. The long-term effect of bed bake-outs on LANA75 material, and how long the benefits last, are unknown and will continue to be monitored. Performing bake-outs results in partially regenerating a LANA75 hydride bed and has several potential benefits.

One benefit is the recovery of tritium and ^3He , both valuable resources. Tritium is eventually recovered from LANA75 hydride beds by performing a time-intensive isotopic exchange process¹² at the end of their service life. A bake-out can be performed any time to recover some of the trapped tritium. There is currently no process in place to recover ^3He heels from postservice hydride beds. Performing occasional bake-outs can recover a portion of this ^3He heel as needed.

Another benefit is clearly realized in routine operations. The improved capacity benefits operations and allows larger quantities of gas to be delivered at required purities. A third benefit of performing bake-outs is that the service life of a bed might be increased, leading to a reduction in the frequency of time-intensive and costly outages necessary to replace beds.

Performing a bake-out to partially regenerate hydride beds requires minimal effort and duration. The results of the bake-out are promising and provide data for future bed development and design. Future bed designs should incorporate heaters and materials that can allow a full regeneration. This would improve routine operations and could significantly reduce the frequency of expensive bed replacement outages, as well as allow for in-process recovery of ^3He . Additional bake-outs should also be performed to partially regenerate additional beds.

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