Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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THERMAL ENHANCEMENT CARTRIDGE HEATER MODIFIED (TECH MOD) TRITIUM HYDRIDE BED DEVELOPMENT. PART II – EXPERIMENTAL VALIDATION OF KEY CONCEPTUAL DESIGN FEATURES

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The Thermal Enhancement Cartridge Heater Modified (TECH Mod) tritium hydride bed is an interim replacement for the 1st generation (Gen 1) process hydride beds currently in service in the Savannah River Site (SRS) Tritium Facilities. Three new features are implemented in the TECH Mod hydride bed prototype: internal electric cartridge heaters, porous divider plates, and copper foam discs. These modifications will enhance bed performance and reduce costs by improving bed activation and installation processes, in-bed accountability measurements, end-of-life bed removal, and He-3 recovery.

A full-scale hydride bed test station was constructed at the Savannah River National Laboratory (SRNL) in order to evaluate the performance of the prototype TECH Mod hydride bed. Controlled hydrogen (H_2) absorption/desorption experiments were conducted to validate that the conceptual design changes have no adverse effects on the gas transfer kinetics or H_2 storage/release properties compared to those of the Gen 1 bed. Inert gas expansions before, during, and after H_2 flow tests were used to monitor changes in gas transfer rates with repeated hydriding/dehydriding of the hydride material. The gas flow rates significantly decreased after initial hydriding of the material; however, minimal changes were observed after repeated cycling. The data presented herein confirm that the TECH Mod hydride bed would be a suitable replacement for the Gen 1 bed with the added enhancements expected from the advanced design features.

I. INTRODUCTION

The Savannah River Site (SRS) Tritium Facilities currently uses 1st generation (Gen 1) $LaNi_{4.25}Al_{0.75}$, or LANA0.75, metal hydride storage beds for tritium storage and separation.[1,2] These beds absorb and desorb hydrogen isotopes by thermal cycling using a hot and cold nitrogen system that is costly and difficult to maintain. Later generation bed designs, 2nd generation (Gen 2) and 3rd generation (Gen 3) beds, have been developed that eliminate the need for a hot/cold nitrogen system and demonstrate additional performance enhancing features.[3,4] The Gen 3 bed is the ideal choice for the upcoming hydride bed replacement in the SRS Tritium Facilities; however, the characterization and performance analysis of this bed still needs further development. Thus, an interim replacement for the Gen 1 bed that can be retrofitted into the current operating configuration, and require minimal testing, was developed.

The Thermal Enhancement Cartridge Heater Modified (TECH Mod) hydride bed boasts the same dimensions and hydrogen storage capacity as the Gen 1 bed, but incorporates several of the enhanced features present in the Gen 2 and Gen 3 beds. The 3 key design modifications in the TECH Mod bed include (1) internal electric cartridge heaters, (2) porous divider plates, and (3) copper heat transfer foam. These features will significantly reduce time, labor, and costs associated with the activation, installation, maintenance, and removal of these beds in the SRS Tritium Facilities.

The internal heaters will improve the end-of life process by allowing the vessels to be heated above the limit of the hot nitrogen system. At higher desorption temperatures (and pressures), more gas will be removed with the same vacuum pumping capabilities. With less residual gas left on the bed with each heating cycle, fewer isotopic exchanges will be required and thus reduce the gas volume needed for this process. The electric heaters could also simulate tritium decay for in-bed accountability (IBA) measurements. This would greatly reduce the time, tracking, and measurement requirements for full bed inventories of tritium. Lastly, the ability to heat the bed to much higher temperatures will facilitate He-3 release and thus extend the life of the hydride bed.

The filter plates within the bed will compartmentalize the hydride material to ensure that an equal mass of hydride material and void space for material expansion is present in each zone of the vessel. The plates are porous to allow sufficient gas flow across the length of the vessel while keeping the hydride particles from migrating between zones. The hydride material is embedded (supported) in a copper foam material to prevent movement of the particles during transport and operation, eliminate the need for manual hydride material leveling, and improve heat transfer within the bed. The combination of these three design features also makes remote activation an attractive possibility. The electric heaters will allow for activation of the hydride bed outside of the Tritium Facilities, while the filter plates and copper will foam prevent the hydride material from moving during post-activation transport and glovebox installation.

Before the TECH Mod hydride bed can be deployed in the Tritium Facilities, flow characterization testing was first performed in the laboratory to verify that the modified design operates within the same characteristics as the Gen 1 bed. Herein, we describe the activation and flow characterization of the prototype TECH Mod hydride bed using a full-scale hydride bed test station designed and constructed here at SRNL. The testing was intended to confirm that not only is the TECH Mod hydride bed a suitable replacement for the Gen 1 beds, but that its enhanced design features will lead to significant process improvements.

II. EXPERIMENTAL

The following experiments were designed to confirm that the TECH Mod design changes have no adverse effects on the hydride bed performance, ensure that gas flow is not significantly affected after several hydriding/dehydriding cycles as a result of decrepitation and/or movement of the hydride material, and evaluate the hydriding properties of LANA0.75 in a full-scale hydride bed. A schematic drawing of the TECH Mod hydride bed, including internal thermocouple (TC) measurement points, is shown in Figure 1. Details of the design and fabrication of the prototype TECH Mod hydride bed are described in a separate manuscript.[5] All testing was performed on a full-scale hydride bed test manifold designed and constructed at SRNL.

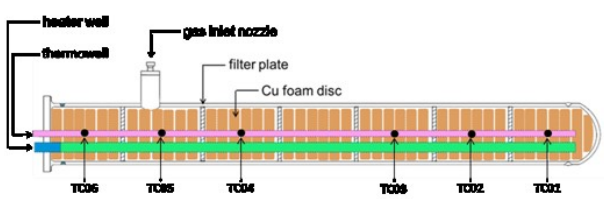

Figure 1. Schematic drawing of the prototype TECH Mod hydride bed.

Inert gas flow characterization (pre- and post-hydriding). Prior to activation of the TECH Mod hydride bed, a series of inert gas expansions using Ar was performed in both the Gen 1 and TECH Mod hydride beds at room temperature. Calibrated volumes (CV) of 1 L and 10 L were used for the baseline flow measurements with initial Ar pressures of 35, 70, 103, 138, 276, 414, and 690 kPa. The pressure equilibration times were measured for each condition (CV→bed and bed→CV) and used as the baseline flow performance for the bed. The pressure curves for each expansion were normalized by dividing by the initial pressure, P_i, in the CV or bed. The inert flow characterization of the TECH Mod hydride bed was repeated after every 4 hydriding/dehydriding cycles for comparison to the baseline measurements. The post-hydriding inert expansions were performed as described for the baseline measurements, but only with the 10 L CV and initial Ar pressures of 35, 70, 103, and 138 kPa.

TECH Mod hydride bed activation. After the baseline flow characterization, the LANA0.75 (12.65 kg) loaded into the TECH Mod bed was activated (or conditioned) to ensure that the maximum amount of H_2 would be absorbed during later tests. An initial bakeout of the bed at 150°C under vacuum was first performed to remove any impurities (i.e., water) in the vessel and hydride material. Next, a series of room temperature H_2 absorption/desorption cycles was performed until the full capacity of the bed was consistently achieved (~0.7 H/M, ~1500 STP-L, 3 cycles performed). Sampling of the off-gases during desorption by residual gas analysis (RGA) was conducted to detect any trace gases. The RGA was calibrated using pure H_2, N_2, Ar, and CH_4 to obtain sensitivity curves and determine the optimal operating pressure for gas sampling.

Absorption/desorption cycling with hydrogen gas. Hydrogen flow characterization tests were performed at 3 different initial bed temperatures: RT (~24°C), 80°C, and 120°C. At each temperature, 4-5 complete H_2 absorption/desorption cycles were conducted. For absorption tests, the maximum amount of H_2 was absorbed onto the bed at initial loading pressures of 933, 733, or 267 kPa. Absorption of H_2 at a constant rate of 2 SLPM (standard liters per minute) was also tested at each temperature. Controlled desorptions at flow rates of 2, 4, and 6 SLPM (80°C and 120°C only) were performed in order to remove the H_2 from the bed. The gas was desorbed into all 7 CVs on the manifold which were open directly to the vacuum pumps. The temperature of the bed was adjusted manually in order to maintain enough pressure in the bed to desorb at the desired flow rate for as long as possible. Once the flow rate could no longer be maintained and most of the H_2 had been desorbed, the bed was baked out overnight at 150°C, followed by cool down to the initial bed temperature.

III. RESULTS AND DISCUSSION

Baseline Flow Characterization. Inert gas (Ar) flow in both the Gen 1 and TECH Mod hydride beds was evaluated in order to (1) confirm that the design modifications of the TECH Mod hydride bed have no significant impact on gas flow compared to the Gen 1 bed and (2) create a baseline comparison for post-hydriding inert flow characterization. The expansion of Ar from CV→bed is referred to as an "absorption", whereas the reverse expansion from bed→CV is described as a "desorption". As expected, the "absorption" equilibration time noticeably decreased as the initial pressure of the gas was increased. Overlays of the "absorption" and "desorption" curves for the Gen 1 and TECH Mod hydride beds indicate comparable flow performance and

equilibration rates for a given starting pressure. The deviation at the equilibrium pressure during the "absorption" processes is a result of the slight difference in void volume (and transfer line volume) of the two beds. Thus, the baseline flow results indicate that the new design features of the TECH Mod hydride bed should not hider gas flow.

Hydrogen Flow Characterization. The main concern of the added features of the TECH Mod hydride bed is that, after repeated cycling of the bed, the decrepitated LANA0.75 material will block the filter plates and/or gas inlet nozzle. This could potentially restrict gas flow through the vessel, leading to major performance and safety/pressure protection issues. Thus, a series of H_2 absorption/desorption cycles was performed on the activated TECH Mod hydride bed to abate these concerns.

Absorptions. For each absorption cycle, H_2 gas was expanded into the hydride bed (at various loading pressures or a fixed flow rate) until maximum absorption was achieved at a given bed temperature and loading pressure. The pressure inside the vessel was monitored over time and allowed to equilibrate for at least 18 h. For example, the data collected at 933 kPa loading pressure are plotted in Figure 2 as the amount of H_2 (STP-L) absorbed in the TECH Mod bed over time. The initial bed temperature and the total amount of H_2 loaded into the system are shown in the figure legend.

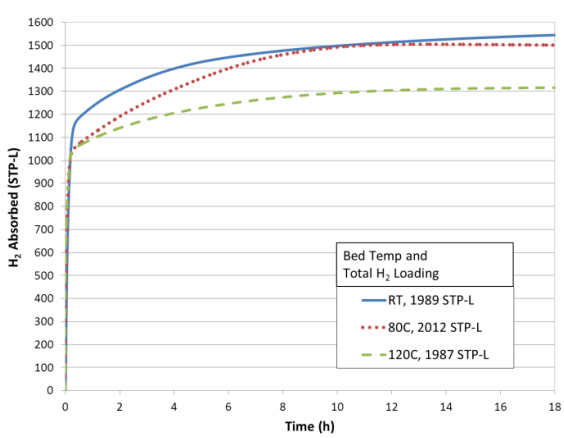

Figure 2. Amount of H_2 absorbed over time in TECH Mod hydride bed (LANA.75) at different bed temperatures and 933 kPa initial loading pressure.

Regardless of the starting bed temperature, the majority of the gas was absorbed in the first few seconds of the expansion. After ~10-12 h, the expected ~1500 STP-L H_2 capacity was reached for the RT and 80°C bed temperatures whereas only ~1300 STP-L were absorbed for the 120°C bed temperature despite the same amount of H_2 present in the system (~2000 STP-L). This lower H_2 loading at 120°C is attributed to the increase in equilibrium pressure of the plateau region of the LANA0.75 isotherm with increasing temperature. Essentially, as the temperature of the bed increases, the equilibrium pressure of the gas above the hydride material also increases; therefore, the hydride material cannot absorb more gas if the loading pressure is at or below that of the equilibrium pressure.

Similar results were obtained for the absorptions at 733 and 267 kPa H_2 loading pressures; however, the 1500 STP-L capacity was never quite reached (~1000-1400 STP-L) as a result of the LANA0.75 equilibrium pressure considerations, as described above. Additionally, because the volume of the test system is limited, multiple aliquots of gas were often required at the lower loading pressures to achieve maximum H_2 absorption in the hydride bed. This was also the case in the 2 SLPM fixed rate absorption in which recharging of the CVs was required so as not to exceed the maximum pressure differential allowed by the flow controller.

Desorptions. After each absorption cycle, the H_2 gas was removed by a controlled desorption experiment. Desorptions at fixed flow rates of 2, 4, and 6 SLPM were performed, in addition to variable flow experiments to determine the maximum desorption flow rates of the TECH Mod hydride bed. For the fixed flow rate desorption, depending on the total amount of H_2 absorbed on the bed, the flow rates were steady and easily maintained for up to 12 h (an example of a 2 SLPM desorption is shown in Figure 3). Additionally, by controlling the desorption flow rate, the bed was able to be desorbed at temperatures below the bed bakeout temperature (~120-130°C as opposed to ~150°C).

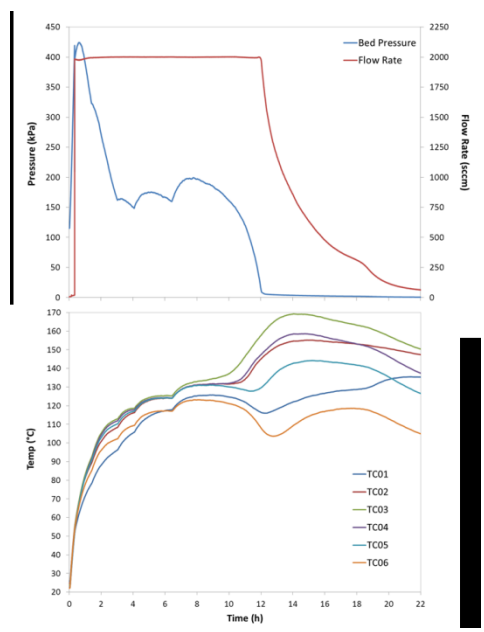

Figure 3. TECH Mod hydride bed desorption at a flow rate of 2 SLPM and RT initial bed temperature.

Post-Hydriding Flow Characterization. During the H_2 flow characterization of the TECH Mod bed, Ar expansions were repeated after every 4 hydriding cycles, where 1 cycle refers to a full H_2 absorption and desorption. Expansions to and from the 10 L CV at initial Ar pressures of 35, 70, 103, and 138 kPa were obtained after 4, 12, and 16 hydriding cycles for comparison to the baseline measurements. The post-hydriding inert flow measurements followed the same trends as the baseline flow data in that the equilibration times decreased with increasing starting pressure. However, there is a marked difference in the inert gas flow before and after the introduction of H_2 gas to the hydride bed. For example, the equilibration times after 12 hydriding cycles of the TECH Mod hydride bed increased from 1-3 min to 12-16 min (>30 min for the 35 kPa test) for the "absorptions", and from 2-6 min to 20-45 min for the "desorptions".

The reduced gas flow rate after hydriding does not necessarily indicate blockage of the filter plates. In fact, the slowed kinetics is more likely a result of decreased Ar flow rates through the finer particles of hydride material (decrepitated LANA0.75), expected after repeated H_2 cycling. If movement of the hydride material and ultimate blockage of the filter plates and/or gas inlet nozzle was restricting the Ar flow, the equilibration times would continue to increase with each hydriding cycle as the LANA0.75 continued to decrepitate. However, this was not observed in the TECH Mod hydride bed. Figure 4 shows an overlay of the 103 kPa Ar expansions from a 10 L CV into the hydride bed after 0, 4, 12, and 16 hydriding cycles. Despite a significant increase in the equilibration time after the initial hydriding (0 cycles, baseline), minimal changes were observed after 4, 12, and 16 cycles. Similar results were also achieved at Ar pressures of 35, 70, and 138 kPa.

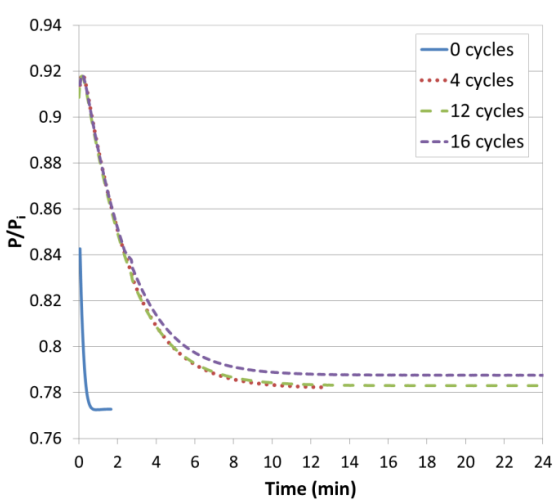

Figure 4. Ar expansions (103 kPa) from 10 L CV into evacuated TECH Mod hydride bed after 0, 4, 12, and 16 hydriding cycles.

IV. CONCLUSIONS

The flow characterization of the prototype TECH Mod tritium hydride bed has been completed. Hydrogen absorption/desorption cycling as well as pre- and post-hydriding inert gas flow measurements confirmed that the advanced design features of the TECH Mod bed sufficiently immobilized the hydride material without inducing any adverse effects on gas flow or bed performance. This work has successfully (1) demonstrated the TECH Mod bed as a viable replacement for the Gen 1 beds currently used in the SRS Tritium Facilities and (2) validated several key design modifications to be implemented in next generation hydride beds. These enhanced features will improve bed performance while significantly reducing costs by eliminating the hot/cold nitrogen system and facilitating bed activation, installation, maintenance, tritium accountability/tracking, end-of-life bed removal, and potentially He-3 recovery.

ACKNOWLEDGMENTS

The authors would like to thank James Klein, Edwin Estochen, Henry Sessions, Devin Staack, Behzad Torkian, and Yanina Breakiron for their contributions to this work. This manuscript has been authored by Savannah River Nuclear Solutions, LLC under contract No. DEAC09-08SR22470 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting this article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for United States Government purposes.

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