DEVELOPMENT OF A PASSIVELY COOLED, ELECTRICALLY HEATED HYDRIDE (PACE) BED

J. E. Klein
Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808
(803) 725-4203

J. R. Brenner
Florida Institute of Technology
150 West University Blvd.
Melbourne, FL 32901
(407) 674-7560

E. F. Dyer
Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808
(803) 725-4624

ABSTRACT

A nominal 1500 STP-L PAssively Cooled, Electrically heated hydride (PACE) Bed has been developed and tested. The bed contained 12.6 kg of a La-Ni-Al alloy and used aluminum foam to improve heat transfer within the bed. Steady-state temperature measurements made at constant power showed a non-uniform bed temperature profile. Protium absorption rates were measured at pressures of 253 kPa, 413 kPa, and 680 kPa with forced convection cooling air flow rates ranging from 50 to 150 SLPM air. Absorption tests were also performed simulating the absorption of tritium and a method for estimating this rate using protium absorption data presented. Desorption rates were measured at pressures ranging from 20 kPa to 933 kPa using dual and single 400 watt electric heaters and found desorption rates were only impacted at the beginning and the end of a desorption cycle by the use of a single heater.

I. INTRODUCTION

The Savannah River Site (SRS) Tritium Facilities currently use 12.6 kg hydride beds to store tritium. The beds desorb and absorb hydrogen isotopes by a thermal swing using large flows of hot and cold nitrogen through the jacket of a bed.[1,2] It was desired to develop an alternate style tritium metal hydride storage bed that does not rely on using hot and cold nitrogen as a replacement for the existing storage beds.

The PAssively Cooled, Electrically heated hydride (PACE) Bed was chosen for development and testing. The prototype bed incorporated two new features not present in existing SRS tritium storage beds. One feature was the use of aluminum foam inside the bed to increase the bed's internal thermal conductivity.[3] The second feature was the use of divider plates to compartmentalized the metal hydride into sections to prevent large scale movement of the material within the bed[3] to prevent excessive hydriding wall stresses.

Passive cooling for the bed was to be by natural convection flow of the glove box atmosphere through the annular space between the bed process vessel (PV) and its external/vacuum jacket with heat rejection to the glove box atmosphere. The vacuum jacket would also provide insulation during desorption to reduce heater power requirements and the external surface temperature of the jacket. To evaluate faster absorption rates for the PACE Bed, bed absorptions were tested using moderate rate forced convection flow through the vacuum jacket.

The In-Bed Accountability (IBA) tritium measurement technique has been developed for inventory measurements without removing tritium from the existing SRS metal hydride storage bed[4,5,6] by passing a constant gas flow through the external jacket of the bed and correlating the steady-state temperature rise of the gas with power. An internal U-tube was incorporated into the design of the PACE Bed to perform IBA measurements with test results to be presented in a future publication.

This paper presents measured absorption and desorption rates of the PACE Bed for its potential use as a replacement for beds continuously cycled using hot and cold nitrogen. Protium absorption rates were measured at different pressures and cooling air flow rates. Electric heaters were used for desorption tests and to simulate tritium to determine its effect on absorption rate. Techniques are presented showing how measured absorption rates can be used to estimate absorption rates with a bed filled with a different metal hydride and how tritium absorption rates can be estimated using protium absorption rate data.

II. PACE BED DESCRIPTION

The PV was made from 7.62 cm (three inch) Sch40 stainless steel (SS) pipe and was approximately 117 cm (46 inches) long. Six, 9.53mm (0.375 inch) OD tubes penetrated through the "header" end of the PV, but did not penetrate the other, "butt", end of the PV. Ten pieces of

Duocell[7] open-cell aluminum foam were installed inside the PV and were separated by nine aluminum divider plates to minimize hydride movement within the PV.

A thermowell in the center of the PV held a three element thermocouple (TC) that measured temperatures at the first (header end) divider plate, the center of the bed, and the last (butt end) divider plate. The other five tubes were centered at various positions on a 2.54 cm (one inch) radius from the center of the PV. A 5 μm sintered metal filter tube was used as the gas process tube and was located above the fill level of the metal hydride. Two heaterwells each held a 400 watt heater with an internal TC located approximately at the mid-point of the heated section. A "U-tube", a single tube with one 180° bend, had the final two penetrations through the header end cap. The U-tube was to supply IBA coolant flow for simulated tritium measurement tests, but could also be used to supply additional cooling to the bed.

Ergenics[8] HY-STOR™ 210, $LaNi_{4.25}Al_{0.75}$ (LANA.75), metal hydride was the desired material for the bed. 12.60 kg of metal hydride was used in the PACE Bed, but isotherm measurements, discussed below, showed PACE LANA properties closer to a previously analyzed $LaNi_{4.70}Al_{0.30}$ (LANA.3) material. A molecular weight corresponding to LANA.3, 422.9 g/mole, was used for PACE LANA protium-to-metal (H/M) atom ratio calculations.

The bed's vacuum jacket was made from 10.2 cm (four inch) Sch10 SS pipe fitted with two, 15.2 cm (six inch) ConFlat® flanges. Two surface TCs were attached to the exterior surface of the jacket (skin) at the center of the bed: one on top and one on the bottom. Inlet and outlet jacket air temperatures were measured using TCs with their tips positioned to the same elevation as the ConFlat® flange gaskets. Figure 1 shows a x-ray of the header end of the assembled bed.

III. EXPERIMENTAL

A. Isotherms

Protium absorption and desorption isotherms on a five gram PACE LANA hydride sample were measured at 30 and 90°C using pressure-volume-temperature measurements. A van't Hoff plot of absorption pressures, Ln[P(kPa)/101.3] versus reciprocal temperature, gave the heat (ΔH) and entropy (ΔS) of formation at 0.4 H/M of -34.1 kJ (-8.14 kcal) per mole-H_2 and -107.3 J (-25.65 cal) per mole-H_2-K, respectively, for the PACE LANA compared to ΔH of -41.4 kJ (-9.89 kcal) per mole-H_2, ΔS of -111 J (-26.6 cal) per mole-H_2 for a LANA.75 material and ΔH of -34.3 kJ (-8.20 kcal) per mole-H_2, ΔS of -110 J (-26.4 cal) per mole-H_2 for a LANA.3 material.

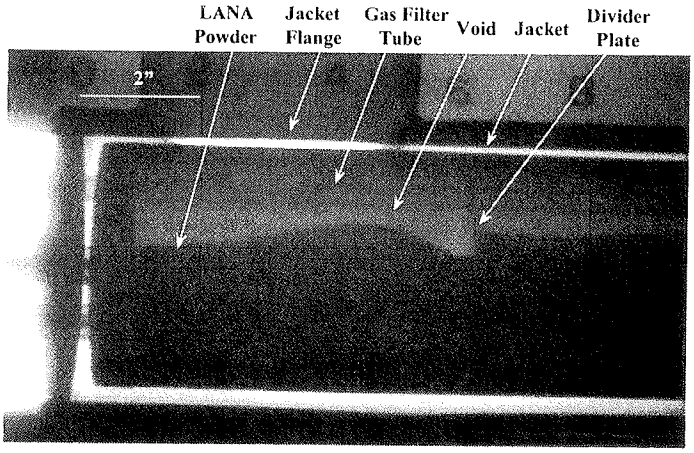

Figure 1. Assembled PACE Bed Header End X-Ray

B. Gas Handling Manifold

Industrial grade hydrogen (protium), minimum purity of 99.95%, was used for all tests. Figure 2 is a simplified test manifold schematic. Pressure was controlled by pressure regulators or pressure control valves. Flows were measured using thermal by-pass mass flow controllers (MFCs) set at their full scale set points and total gas transfers were calculated by integrating flow-time data. Figure 2 shows separate MFCs for absorption and desorption measurements, but manifold valving not shown utilized a single MFC for all hydrogen flow measurements. PT in Figure 2 indicates single or multiple capacitance manometers and/or thermocouple vacuum gauges.

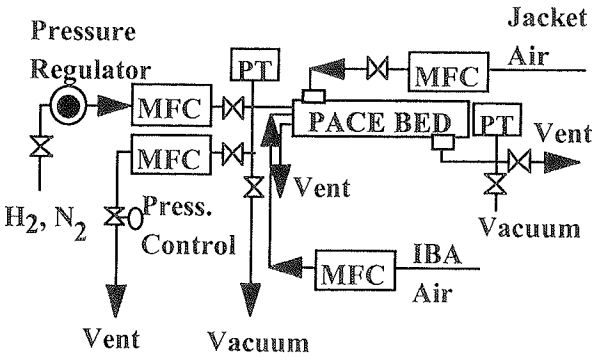

Figure 2. Schematic of PACE Bed Test Manifold

C. Bed Activation

The bed was evacuated using a two-stage oil pump, heated under vacuum to 150°C, then exposed to protium at a maximum of 253 kPa. After partial activation, the bed was desorbed at 150°C under vacuum, cooled to ambient, and again re-exposed to protium. This activation process was repeated several times until the hydride would readily absorb protium.

D. Testing

Absorption tests were run at 253 kPa, 413 kPa, and 680 kPa which correspond to isotherm equilibrium temperatures of 68.4°C, 83.0°C, and 99.1°C, and LANA.75 absorption pressures of 30.9 kPa, 56.3 kPa, and 103 kPa, respectively. Absorption rate tests above 680 kPa were not possible due to apparatus limitations so rate estimates at higher pressures/temperatures for the desired LANA.75 hydride were made by data extrapolation.

For absorption tests, ambient air was supplied by an MFC to the bed jacket. Unless stated otherwise, air flow was from the header end to the butt end of the bed. A "cold" start absorption refers to a test where the bed was initially at ambient temperature. A "hot" start absorption refers to the bed at a temperature that would generate the absorption test pressure. The protium pressure to the bed was controlled by a pressure regulator and measured using a MFC.

The bed heaters were used and power settings manually adjusted for tests simulating the absorption of 100% tritium. The heater power was set to estimate the decay heat of the residual tritium in the bed before the start of an absorption test. After approximately every additional 100 STP-L of protium absorbed by the bed, the heater power was increased appropriately to simulate the tritium decay heat. For all tests simulating tritium decay heat, the heater power levels were set to correspond to 1.95 watts/mole of tritium.

For desorption tests, the bed jacket was evacuated to less than 40 Pa and the tests started near ambient conditions. Protium evolution from the bed was controlled by a pressure controller and measured using a MFC. Heater power levels were adjusted manually as needed during the run to generate the desired desorption pressure, flow rate, simulated decay heat, or to prevent the bed from exceeding 150°C at any heater well TC location.

IV. RESULTS

A. PV Temperature Profiles

Steady-state PV hydride temperature profiles at a nominal 127 watts internal power with different vacuum jacket cooling rates showed that under full vacuum the center TC was 151°C. Under all conditions, the center TC was approximately 40°C hotter than the header TC. At flows less than 25 SLPM, the center of the bed was the hottest and the header end the coolest due to the six tubes penetrating the header end cap acting as cooling fins. With air flows of 25 SLPM or greater through the jacket, the butt end was the hottest due to the heating of the air as it flowed through the jacket. Reversing the air flow direction essentially exchanged the header and butt temperatures although, some slight differences were obtained.

B. Absorption Tests

Initial PACE Bed tests compared cold versus hot start conditions at a protium absorption pressure of 253 kPa and a cooling rate of 100 SLPM. The initial cold start absorption rate was over 20 SLPM, decreased, and then followed a somewhat regular decreasing rate. The hot start absorption rate was low after the initial gas filling of the bed void volume, increased, and then followed a trend similar in decreasing absorption rate as was found with the cold start absorption. The absorption rate at the time to absorb 1500 STP-L, the 1500L-Rate, occurred at 7 hours for a cold start test and 11.3 hours for a hot start test.

Hot start absorption testing was chosen over cold start absorption testing to measure worse-case absorption rates and rates that may be typical of a process operation where beds could be cycled frequently. Figure 3 shows hot start absorption rates as a function of time for different pressures and cooling air flow rates. The 1500L-Rates are shown as vertical tick marks on the rate lines in Figure 3.

Figure 4 shows the 1500L-Rate data from Figure 3 versus equilibrium absorption temperature. 1500L-rates corresponding to LANA.75 absorptions at 66.7 kPa and 253 kPa, 87.4 and 126°C respectively, can be estimated from Figure 4 by extrapolation of the rates (dashed lines) from the lower temperature data.

C. Absorption Tests With Simulated Decay Heat

Hot start absorption tests simulating tritium absorption at 680 kPa and 413 kPa with a cooling flow rate of 100 SLPM showed a jagged stair-step shaped absorption curves of decreasing absorption rate due to the increase in heater power simulating the absorbed tritium.

The quick decrease in absorption rate following an increase in heater power illustrated the effectiveness of the aluminum foam in enhancing internal heat transfer in the bed.

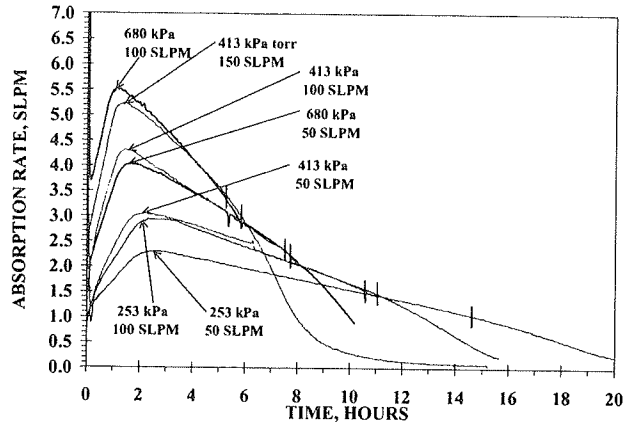

Figure 3. Hot Start Absorption Rates

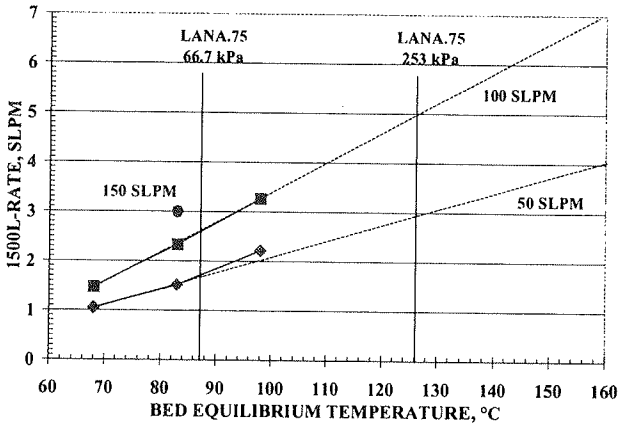

Figure 4. Hot Start Rate Estimates for LANA.75 Metal Hydride

Absorption rates for tritium can be estimated using protium absorption data (without heaters), the tritium decay heat, and the hydride heat of formation by first equating the amount of protium in the bed to tritium and calculating the tritium decay power using 1.95 watts per mole tritium. Then divide the decay heat power by the hydride heat of reaction (ΔH) to generate an equivalent desorption rate of gas due to the tritium decay heat (i.e. a reduction in absorption rate) and subtract this desorption rate from the experimentally measured absorption rate. As additional gas is absorbed by the bed, recalculate the net absorption rate.

Figure 5 shows hot start protium absorption data without heater power ("0%" T_2 data), hot start simulated tritium absorption data ("100%" T_2 data), and the calculated tritium absorption rates using the protium absorption results at 680 kPa and 413 kPa. The calculated absorption rates at 680 kPa in Figure 5 were shifted a positive 0.06 H/M to compensate for the differences in the residual gas in the bed before the start of the tests.

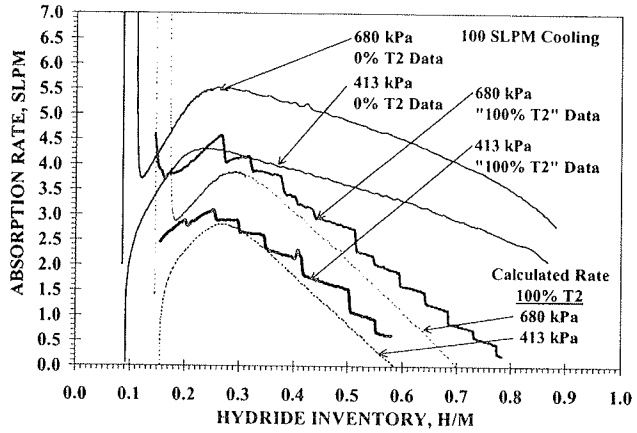

Figure 5. Calculated Tritium Absorption Rates

A test was run to measure the effect of "inerts", non-hydriding gas, had on absorption rate. Nitrogen (99.9995% purity) was introduced into the bed at two discrete times during a hot start absorption test at 680 kPa, 100 SLPM cooling, while simulating the absorption of tritium. At the start of the test, 0.57 STP-L of nitrogen was introduced into the bed. Approximately 1.5 hours into the test, the absorption was stopped, an additional 0.75 STP-L of nitrogen added to the bed, and the absorption test continued. The addition of the 1.32 STP-L of nitrogen to the bed had virtually no impact on the bed absorption rate when compared to tests without the addition of nitrogen.

D. Desorption Tests

Figure 6 shows the results of 625 watt desorption tests controlled at pressures of 200 kPa, 413 kPa, and 547 to 640 kPa. The desorption flow rate for the 200 kPa desorption was limited during part of the test by the use of a 20 SLPM full-scale range MFC. Heater power was held constant until the bed center TC or the butt TC approached 150°C after which the power was manually adjusted to maintain a 150°C hydride temperature until the end of the desorption test.

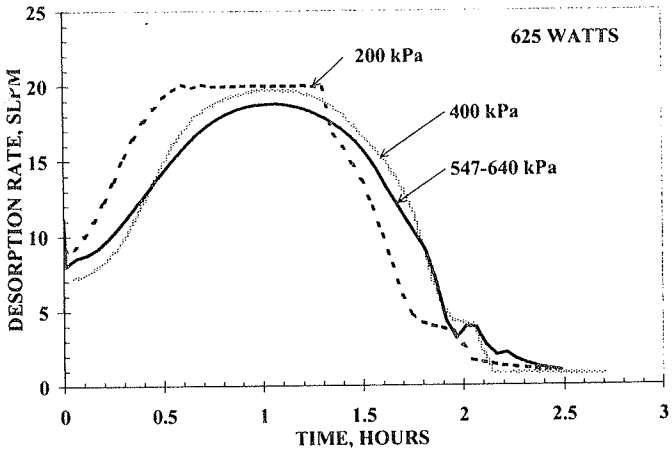

Figure 6. Constant Power Desorption Rates

It was noticed after the desorption tests less than two minutes were needed for the heater TCs to equilibrate with the bed TC temperatures after the heater power was discontinued showing the rapid thermal equilibration of the bed in the absence of hydriding.

Variable power desorption tests were tried with both, and then one heater, where the target desorption pressure was 933 kPa and the target desorption rate was six SLPM. Initially, more than 400 watts were needed to heat the bed and to deliver the gas at the test conditions during the dual heater test. After less than 0.5 hours, the power needs were reduced to less than 400 watts to maintain the test conditions and were constant at 260 watts for over two hours. After this time, the power was increased to maintain test conditions. After 4.3 hours, the bed center TC reached 150°C and the power was reduced to prevent bed overheating with a subsequent decrease in bed delivery pressure.

The single heater desorption test was at full power at the start of the test and generated the target pressure, but a flow rate below target conditions until approximately one hour into the test. After one hour, the power required to maintain test conditions was nearly constant at 260 watts, the same power level as during the dual heater test, for two hours. After two hours at approximately 260 watts, the heater power was increased and eventually reached its 400 watt maximum which then caused a decrease in desorption rate.

V. DISCUSSION

Cold start bed absorptions have the obvious advantage of absorbing more gas in less time than with a hot start bed. Cold versus hot start tests showed that after the initial transients pass, the slope of the absorption rate-time data was nearly constant for several hours, independent of whether the bed was hot or cold at the start of the test. During absorptions when the change in absorption rate was nearly constant, an energy balance shows the heat released from the hydriding reaction, the absorption rate times ΔH, was balanced by the heat carried away by the cooling gas flow and natural convection losses when using a heat transfer coefficient of 6.24 W/m^2-°C (1.10 BTU/hr-ft^2-°F), the jacket's geometric area, and the difference in the average of two external jacket ("skin") TC temperatures and the surrounding lab temperature.

The near matching of absorption rates under different pressure/flow conditions was shown in Figure 3. The difference in equilibrium absorption temperatures corresponds roughly to 15°C for the test pressures of 680 kPa, 413 kPa, and 253 kPa and the cooling rates tested differed by 50 SLPM. The results show that each additional 50 SLPM of cooling air flow in the range of 50 to 150 SLPM effectively lowers the bed temperature during hydriding by 15°C.

The 1500L-Rate, the hydriding rate after absorbing 1500 STP-L, will determine whether or not the bed can maintain its required hydriding rate at its hydrogen isotope design capacity of 1500 STP-L. The 1500L-Rates extrapolated in Figure 4 are conservative (low) estimates of hydriding rates since at higher temperatures (absorption pressures) the bed would be hotter, have higher heat transfer rates, and thus faster hydriding rates.

Absorption rates for tritium calculated in Figure 5 underestimate those measured with simulated decay heat due to the assumption that all the decay heat goes to desorbing gas (i.e. reducing the absorption rate) instead of also increasing the bed temperature. This makes hydriding rates calculated using non-tritium absorption rate data conservative in that they will underestimate actual absorption rates.

For the absorption test with 1.32 STP-L of nitrogen, the nitrogen occupied 6.6 percent of the PV void volume (2.99 L). After 2.2 hours, the hydriding rate recovered after the second nitrogen additional and a total of 508 STP-L of protium had been absorbed. The 1.32 STP-L of inerts in 508 STP-L of protium is equal to absorbing an inert mixture containing 0.26 percent inerts.

The absorption rate for a 0.26 percent inert mixture with the PACE Bed will not predict the absorption rate for a 0.26 percent inert mixture for a LANA.75 at 103 kPa due to the difference in protium partial pressures during the absorption. For the 1.32 STP-L of nitrogen added during the absorption test, the equivalent inert

concentration for the absorption of 508 STP-L by a LANA.75 material would be 0.0475 percent. To simulate the absorption of 1500 STP-L of protium with 0.1 percent inerts with LANA.75 at 103 kPa , a PACE Bed protium absorption test with 0.552 percent inerts at 680 kPa would need to be done: 8.3 STP-L of nitrogen added to the bed for the test.

The minimum heater power to desorb the bed at approximately 150°C was calculated by adding the bed heat loss power requirements with the bed desorption power requirements. At steady-state conditions, approximately 127 watts were needed to maintain the bed center temperature near 150°C. Desorption of one SLPM of protium requires 25.3 watts for the PACE Bed and 30.8 watts for a LANA.75 bed. For the desorption rate of 20 SLPM shown in Figure 6, 506 of the 625 watts input to the bed were going to desorb the bed with the remaining 109 watts used to increase the temperature of the bed or were lost from the bed. A single 400W heater is sufficient to desorb the bed, but would require more time for heat-up to operating conditions and could suffer reduced flow rates at the beginning and end of a desorption cycle.

VI. CONCLUSIONS

The prototype PACE Bed operating with forced atmosphere cooling has potential for use in SRS or fusion tritium projects. Desorbing gas from the bed is easily accomplished using electric heaters in approximately 2 to 3 hours. Delay times on the order of 1.5 minutes were observed between changes in heater power and rates of hydriding/dehydriding showed the aluminum foam was highly efficient in transferring heat throughout the bed. One disadvantage of the prototype is the long pull length needed, approximately 2.4 meter (eight feet), to replace failed heaters.

Absorption rates for the PACE Bed depend greatly on the cooling air flow rate, absorption pressure, and the amount of tritium in the bed. Absorption of gases containing tritium can significantly reduce the absorption rate of the bed and thus the bed's usable capacity under process conditions. The inclusion of fins on the external surface of the PV, or internal/external fins on the vacuum jacket, may further increase absorption rates. Splitting the cooling flow from all external to the PV to some flow through the vacuum jacket and some through the U-tube has the potential to increase absorption rates with the same total air flow. The bed header penetrations act as cooling fins and create anisotropy in the axial bed temperature profile; this thermal anisotropy is expected to facilitate any maintenance that might become necessary on the header end.

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7. ERG Materials and Aerospace Corporation, 900 Stanford Avenue, Oakland, CA, 94608, U.S.A.

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