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IN-BED ACCOUNTABILITY DEVELOPMENT FOR A PASSIVELY COOLED, ELECTRICALLY HEATED HYDRIDE (PACE) BED

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A nominal 1500 STP-L Passively Cooled, Electrically heated hydride (PACE) Bed has been developed for implementation into a new Savannah River Site tritium project. The 1.2 meter (four-foot) long process vessel contains an internal "U-tube" for tritium In-Bed Accountability (IBA) measurements. IBA will be performed on six, 12.6 kg production metal hydride storage beds.

IBA tests were done on a prototype bed using electric heaters to simulate the radiolytic decay of tritium. Tests had gas flows from 10 to 100 SLPM through the U-tube or 100 SLPM through the bed's vacuum jacket. IBA inventory measurement errors at the 95% confidence level were calculated using the correlation of IBA gas temperature rise, or (hydride) bed temperature rise above ambient temperature, versus simulated tritium inventory.

Prototype bed IBA inventory errors at 100 SLPM were the largest for gas flows through the vacuum jacket: 15.2 grams for the bed temperature rise and 11.5 grams for the gas temperature rise. For a 100 SLPM U-tube flow, the inventory error was 2.5 grams using bed temperature rise and 1.6 grams using gas temperature rise. For 50 to 100 SLPM U-tube flows, the IBA gas temperature rise inventory errors were nominally one to two grams that increased above four grams for flows less than 50 SLPM. For 50 to 100 SLPM U-tube flows, the IBA bed temperature rise inventory errors were greater than the gas temperature rise errors, but similar errors were found for both methods at gas flows of 20, 30, and 40 SLPM.

Electric heater IBA tests were done for six production hydride beds using a 45 SLPM U-tube gas flow. Of the duplicate runs performed on these beds, five of the six beds produced IBA inventory errors of approximately three grams: consistent with results obtained in the laboratory prototype tests.

I. INTRODUCTION

The Savannah River Site (SRS) Tritium Facilities currently use metal hydride storage beds¹ with 12.6 kg of $\text{LaNi}_{4.25}\text{Al}_{0.75}$ for process gas absorption, storage, and desorption. These 1st generation (Gen1) storage beds

contain the metal hydride in a 7.62 cm (3 inch) pipe process vessel surrounded by a 10.2 cm (4 inch) pipe jacket. The beds desorb and absorb hydrogen isotopes by thermal-swing supplied by large flows of hot and cold nitrogen through an insulated jacket.

The In-Bed Accountability (IBA) technique has been developed for tritium accountability (inventory) measurements on the Gen1 storage beds without removal of tritium from the bed.^{2,3,4} For Gen1 beds, the steady-state temperature rise of a constant gas flow through the bed's jacket (ΔT_J^{Gas}) is correlated with tritium content to generate a calibration curve for the bed. ΔT_J^{Gas} is obtained during inventory measurements cycles and the calibration curve used to determine bed tritium content. This technique has been in production use for 10 years.

A 2nd generation (Gen2) metal hydride bed has been developed which does not use hot and cold nitrogen to supply the thermal swing for gas desorption and absorption.⁵ This Gen2 bed, developed as a Passively-Cooled, Electrically heated hydride (PACE) Bed, utilizes electric heaters for desorption and much lower forced atmosphere cooling flow rates than the Gen1 beds for gas absorption. The Gen2 beds will be put into tritium service in 2004 and contain the same mass and hydride alloy composition as the Gen1 beds. These beds are also referred to as Forced-Atmosphere (glove box nitrogen) Cooled, Electrically heated (FACE) Beds.

The ability to measure/determine the amount of tritium in a metal hydride storage bed should be incorporated into the design of the bed and tested as part of the bed's development program.⁶ The purpose of this work was to perform IBA tests on Gen2 storage beds, using electric heaters to simulate tritium decay, and determine the IBA accuracy for this bed design.

II. BACKGROUND

The Gen1 storage bed IBA gas flow is within the jacket of the bed and over the outside of the process vessel. The IBA gas stream is warmed by the tritium in the process vessel, but some heat is transferred and lost through the jacket.

To reduce Gen2 bed IBA heat losses through the jacket, the bed jacket is evacuated and the IBA gas flow through a “U-tube” internal to the process vessel, a concept similar to the internal coil of a ZrCo storage bed.⁶ It was anticipated that lower losses of tritium decay heat through the jacket would improve the accuracy and the detection limit of the IBA technique for Gen2 beds.

High accuracy tritium measurement can be made on metal hydride storage beds,⁷ but simple bed designs are desired for production beds. Incorporation of the U-tube into the bed complicates bed fabrication questioning the need for the U-tube. The Gen2 bed design allows for jacket flow IBA as used for the Gen1 beds. One goal of the test program was to determine IBA gas flow through the U-tube gave superior tritium measurement uncertainty compared to IBA jacket flow. The steady-state IBA temperature rise will be denoted as ΔT_J^{Gas} for jacket gas flow, and ΔT_U^{Gas} for U-tube gas flow. The steady-state IBA temperature rise of the bed, bed thermowell center TC⁵ temperature minus ambient temperature, will be denoted as ΔT_J^{Bed} for jacket gas flow, and ΔT_U^{Bed} for U-tube gas flow.

III. EXPERIMENTAL

A detailed description of the Gen2 prototype bed number 1 (PB1) has been described previously.⁵ Figure 1 is a schematic of PB1 IBA gas flows and thermocouple (TC) placements. The U-tube TC tips were inserted 11.4 cm (4.5 inches) into each leg of the U-tube to correspond to the position of the bed’s 1st divider plate. The jacket TC tips were positioned to be at the same elevation as the ConFlat[®] flange gaskets.

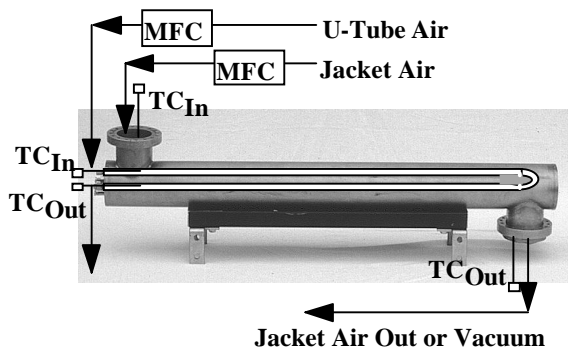


Fig. 1. Schematic of PB1 IBA Test Configuration.

Tritium decay heat was simulated by adjusting the voltage to the bed’s two 400W electric heaters. For U-

tube gas flow, the hydride bed protium pressure was between 22.7-100 kPa (170-750 torr) for 100 SLPM flow and was less than 1.33 kPa (10 torr) for all other U-tube flows. IBA U-tube data were collected at nominal powers of 0, 4.8, 8.6, 25, 43, 59, 75, 92, and 108 watts. For the 100 SLPM jacket flow test, the bed protium pressure was less than 1.33 kPa (10 torr). Data were collected at powers of 0, 25, 48, 73, 96, and 122 watts.

Nominally 24 hours were given to obtain the steady-state data before conditions were varied for the next IBA test. Simulated tritium loadings were expressed as a tritium-to-metal atom ratio, T/M, using 1.95 watts per mole of tritium. A molecular weight of 408.62 g per mole was used in the T/M calculations to better compare results to production beds which use $\text{LaNi}_{4.25}\text{Al}_{0.75}$ instead of the corresponding 422.9 g per mole for the $\text{LaNi}_{4.7}\text{Al}_{0.3}$ in PB1.⁵

Linear and quadratic regression models, with and without zero intercept, were fit to the ΔT versus T/M data to obtain regression coefficients and other statistics parameters. The previously described method⁴ was used to select the “best” regression model at the 95% confidence level and calculate the inverse regression standard deviation, σ_{inv} . The accuracy of the IBA technique was calculated as the product of the statistical student’s t-test value and σ_{inv} : $t^*\sigma_{inv}$. Errors calculated as a percentage of a full bed were based on a bed loading of 0.60 T/M. $t^*\sigma_{inv}$ varies as a function of T/M and the values at 0.6 T/M, the largest values, will be reported.

IV. RESULTS

Figure 2 shows ΔT_U^{Gas} versus T/M for U-tube flows between 10 and 100 SLPM. Similar temperature rise versus T/M relationships (calibration curves) were obtained for the ΔT_U^{Bed} , ΔT_J^{Gas} and ΔT_J^{Bed} data with the jacket flow data showing a larger quadratic dependence of the correlation. At 100 SLPM, the magnitude of the ΔT versus T/M correlations, in decreasing order, were ΔT_J^{Bed} , ΔT_U^{Bed} , ΔT_U^{Gas} , and ΔT_J^{Gas} .

Most “Best” fit models used for inverse regression analyses of the IBA data were quadratic. The $t^*\sigma_{inv}$ error for the four IBA techniques are shown in Figure 3 along with the results for six SRS 1500L storage beds calibrated with tritium.⁴ Smaller $t^*\sigma_{inv}$ errors for U-tube flow versus jacket flow lead to termination of further jacket flow tests.

For a 50 SLPM U-tube gas flow, Figure 4 shows the steady-state bed temperatures relative to ambient at various simulate tritium loadings. It can be seen that the

inlet air temperature TC increases as a function of bed power level and was found to decrease the influence of ambient air temperature fluctuations on the IBA results.

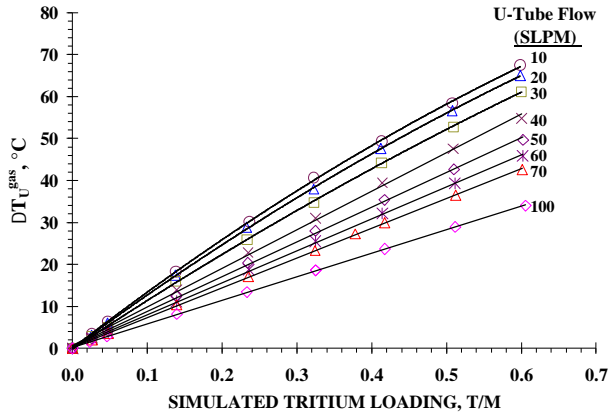


Fig. 2. PB1 IBA U-Tube Gas Flow Temperature Rise.

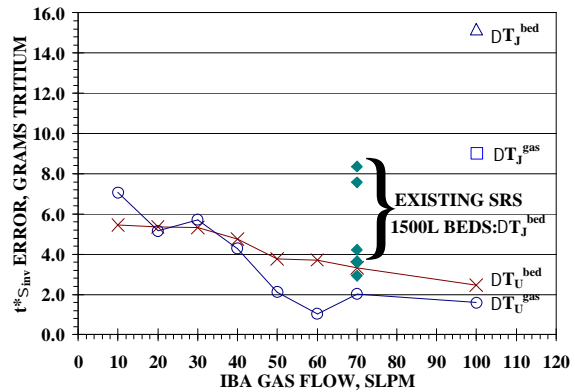


Fig. 3. PB1 IBA Inventory Errors Versus IBA Gas Flow.

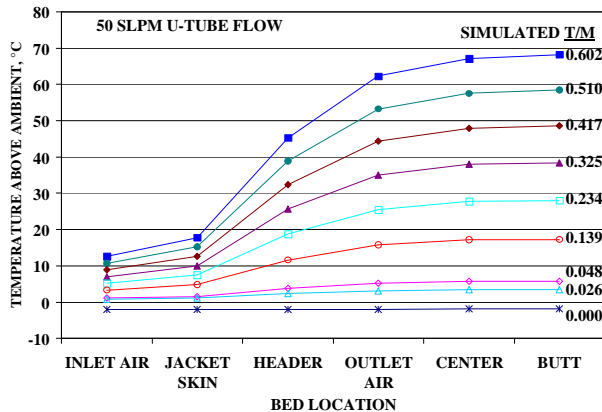


Fig. 4. PB1 Temperatures During IBA.

V. DISCUSSION

Figure 3 shows IBA jacket gas flow gives larger IBA errors than IBA U-tube gas flow. Between 50 and 100 SLPM, the ΔT_U^{Gas} method produced the smallest IBA errors of nominally 2 grams, or 0.60 percent of a full bed. Below 50 SLPM U-tube flow, the ΔT_U^{Gas} and ΔT_U^{Bed} methods produced similar, but larger, IBA measurement uncertainties.

The selection of U-tube gas flow for production bed IBA was balanced by the desire to minimize IBA uncertainties while also minimizing gas consumption. Lower IBA gas flows generate higher bed temperatures and the potential for process tritium permeation emissions during IBA measurements. 45 SLPM was chosen as the production IBA gas flow so “off-the-shelf” 50 SLPM mass flow controllers (MFCs) could be purchased and allow three-point calibration at 40, 45, and 50 SLPM.

Figure 4 illustrates two characteristics of U-tube IBA thermocouple placement. First, the inlet TC temperature correlated linearly with T/M, changing 2.3°C per 0.1 T/M, but was found to be independent of ambient temperature fluctuations. The increase in inlet temperature decreased the absolute magnitude of ΔT_U^{Gas} , but dampened out the influence of ambient temperature fluctuations on inlet gas temperature. The other characteristic was the outlet gas temperature being lower than the bed’s center or butt temperatures. The U-tube gas is cooled below the butt or center temperatures by transferring energy to the cooler/header end of the bed. Higher outlet gas temperatures could have been obtained by deeper insertion of the outlet TC into the U-tube, but was not done in this work.

IBA U-tube gas flow tests were performed at 45 SLPM on six production FACE Beds with the beds filled with 101 kPa (760 torr) helium. The first IBA set, “Run A”, was run at nominal powers of 0, 23, 44, 66, 88, and 110 watts while “Run B” was done at 17, 39, 61, 82, and 104 watts.

Figure 5 shows the IBA ΔT_U^{Gas} $t^*\sigma_{\text{inv}}$ errors for the two production FACE bed runs, the PB1 results, and for the existing RTF beds calibrated with tritium.⁴ The two runs on the production FACE beds were plotted at flows other than 45 SLPM to show the change in error between the two runs.

Figure 5 shows $t^*\sigma_{\text{inv}}$ errors for five of the FACE beds were similar to those obtained for PB1 or the RTF tritium recovery beds run with a 70 SLPM jacket gas flow. $t^*\sigma_{\text{inv}}$ errors greater than five grams for the FACE

beds, especially Bed 090, were typically the result of not waiting for steady-state conditions to be achieved before starting the next test.

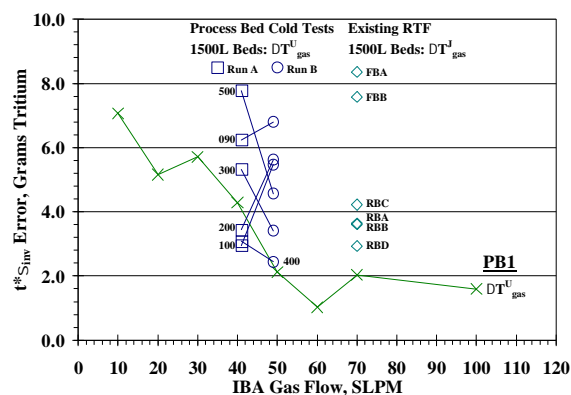


Fig. 5. IBA Inventory Errors for Various Beds.

VI. CONCLUSIONS

IBA gas flow through the U-tube gas gave smaller tritium measurement errors than gas flows through the jacket demonstrating the need for the U-tube. IBA errors are relatively constant for U-tube flows between 50 and 100 SLPM, but increased as the flow decreased below 50 SLPM.

Selection of 45 SLPM production IBA flows allows purchase of commercially available 50 SLPM full-scale range MFCs, three point MFC calibrations by spanning the set point by ± 5 SLPM, and was a reasonable compromise on gas consumption and IBA accuracy.

Improved criteria for determining steady-state conditions for production bed measurements should allow IBA detection limits on the order of three grams per bed (0.97 W) or 0.0054 T/M. For a full bed at 0.6 T/M, the relative error is 0.9 percent.

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REFERENCES

- [1] M. S. Ortman, L. K. Heung, A. Nobile, and R. L. Rabun, "Tritium Processing at the Savannah

River Site: Present and Future," *J. Vac. Sci. Technol. A*, **8** (3), 2881 (1990).

- [2] J. E. Klein, M. K. Mallory, and A. Nobile, Jr., "Tritium Measurement Technique Using 'In-Bed' Calorimetry," *Fusion Techn.*, **21**, 401 (1992).
- [3] J. E. Klein, "In-Bed Accountability of Tritium In Production Scale Metal Hydride Storage Beds," *Fusion Techn.*, **28**, 1026 (1995).
- [4] J. E. Klein, "A Summary of Tritium In-Bed Accountability for 1500 Liter $\text{LaNi}_{4.25}\text{Al}_{0.75}$ Storage Beds," *Fusion Techn.*, **41**, 542 (2002).
- [5] J. E. Klein, J. R. Brenner, and E. F. Dyer, "Development of a Passively Cooled, Electrically Heated Hydride (PACE) Bed," *Fusion Sci. and Technol.*, **41**, 782 (2002).
- [6] T. Hayashi, M. Yamada, T. Suzuki, Y. Matsuda, and K. Okuna, "Tritium Accounting Characteristics of 'In-Bed' Gas Flowing Calorimetry," *Fusion Technol.*, **28**, 1015 (1995).
- [7] J. L. Hemmerich, "Thermal Design of A Metal Hydride Storage Bed, Permitting Tritium Accountancy to 0.1% Resolution and Repeatability," *Fusion Technol.*, **28**, 1732 (1995).