BULK GETTERS FOR TRITIUM STORAGE

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ABSTRACT

A program was carried out to develop working experience with bulk metal getter beds for tritium handling in support of the Tritium Removal Facility being constructed at the Darlington Nuclear Generating Station site. This paper summarizes the data base developed for temporary and long-term tritium storage on bulk getters, namely the operating characteristics of a series of uranium and titanium beds built and tested during this program. Descriptions of doubly contained 3 kg and 25 g uranium beds, recently designed at Ontario Hydro for tritium service, is also presented.

INTRODUCTION

Tritium is produced in significant quantities in the CANada Deuterium Uranium (CANDU) reactors. The Tritium Removal Facility (TRF), which is in the advanced stages of construction at the Darlington Nuclear Generating Station, will extract tritium from tritiated heavy water.(1) The removed tritium will be stored in Immobilization Tritium Containers as titanium "tritide". As demand dictates, tritium will be transferred from the long-term storage titanium beds to temporary storage uranium beds and subsequently metered into appropriate shipping containers.

A research program was formulated to cultivate working experience with bulk getter beds for tritium handling in support of the TRF. This paper is a summary of the data base developed during this program. Specifically, the paper presents the characteristic operating properties of uranium beds of various sizes: simple 15 g bed, doubly contained 6 kg bed, two-tier 660 g bed, and simple 100 g shipping bed. The performance of loading and unloading a simple 200 g titanium bed are also presented. Descriptions of doubly contained 3 kg and 25 g uranium beds, recently designed at Ontario Hydro for tritium service, are given.

URANIUM GETTER BEDS

The low dissociation pressure of uranium hydride at ambient temperature, 30 mPa at 25°C, and a dissociation pressure equal to atmospheric pressure at 432°C (2), essentially independent of composition, make uranium a very versatile getter for tritium service. The narrow operating temperature range of approximately 400°C permits rapid thermal cycling of uranium and thereby making it amenable for a number of swift applications: temporary storage; short, repeated recovery operations; and helium separation from tritium gas. These properties make uranium an ideal material for temporary storage of tritium.

Simple 15 g Bed

The loading and unloading rates for a simple 15 g uranium powder bed were studied on the process loop shown in Figure 1.(3)

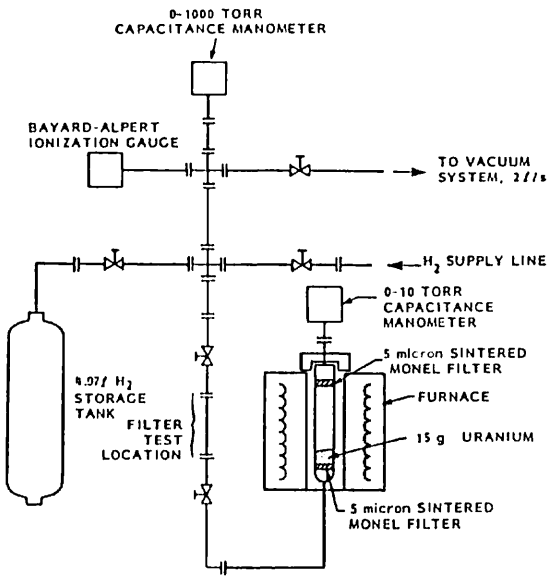

FIGURE 1: 15 g URANIUM TEST BED LOOP

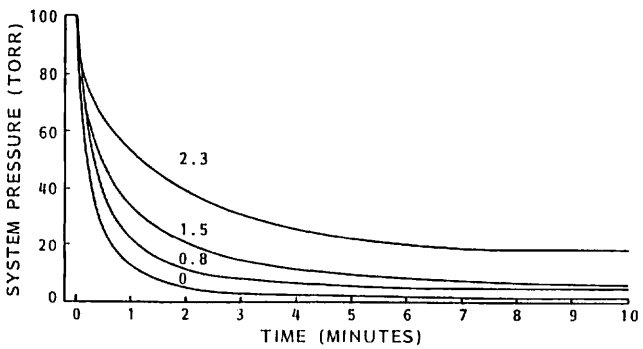

FIGURE 2: DEPENDENCE OF THE LOADING RATE ON THE INITIAL HYDROGEN-TO-METAL RATIO (VALUES GIVEN ON THE CURVES) (FILTER THICKNESS, 0.27 cm; AREA 1 cm^2; POROSITY 5 μm)

The factors influencing the hydriding rate of the bed are the bed over-pressure (bed pressure-equilibrium pressure), the initial bed capacity, the

bed temperature, the bed orientation, the helium content in the gas stream, and the in-line filter porosity. The rate of hydriding increases with increasing bed over-pressure and decreasing initial bed capacity (see Figure 2) and getter temperature. The charging rate is greater for a horizontally oriented bed than that positioned vertically; this is a reflection of greater heat transport from the hydriding region to the outside environment, which is a result of increased contact area between the uranium powder and the surface conducting heat to the external environment. The presence of helium in concentrations of 0.1% or greater noticeably reduces the rate of hydrogen uptake. Placement of filters with increasing tightness (decreasing porosity), for uranium particulate control, decreases the flow conductance and thereby limits the overall hydrogen uptake rate. Therefore, the choice of filter porosity must balance the need to control particulate migration into the loop with the overall rate of hydriding and dehydriding. It is worth noting that the uranium particulates are largely submicron in size and that the filtering action is electrostatic adhesion of the white flecks to the filter surface (see Figure 3).(3)

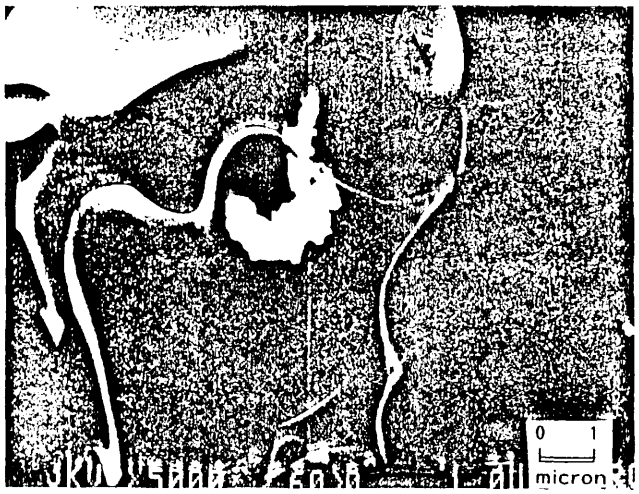

FIGURE 3: TYPICAL URANIUM PARTICULATE TRAPPED ON A 5 μm FILTER

Hydrogen regeneration from the 15 g bed was achieved within 20 min at a temperature of 350°C. A total of 125 hydrogen loadings and unloadings were conducted without any noticeable degradation in the macroscopic performance of the bed.

Doubly Contained 6 kg Bed

The operating characteristics of a doubly contained 6 kg uranium bed, designed and built by Los Alamos Scientific Laboratory (LASL) for the Tritium Systems Test Assembly, were determined at the research laboratories of Ontario Hydro. A schematic of the process loop on which the uranium bed was tested is shown in Figure 4, and an expanded view of the LASL bed is shown in Figure 5.(4)

The factors that influence the loading rate of a large uranium bed, such as the LASL U bed, are those already identified in the previous section. However, given the increase in scale, some of the factors have a greater impact on the performance of the bed. Increase in the quantity of uranium permits more hydrogen loading, implying a greater production of heat. The geometry of the primary vessel vis-a-vis the uranium powder must be optimum to ensure good thermal transport. Furthermore, during loading the secondary volume must be filled with a high thermal conductivity gas for maximum heat transport. In short, the rate of loading is critically dependent on the balance between the rate of thermal transport from the uranium powder to the external environment, via the primary and secondary vessels, and the rate of heat liberation in the uranium powder due to the exothermic hydriding reaction.

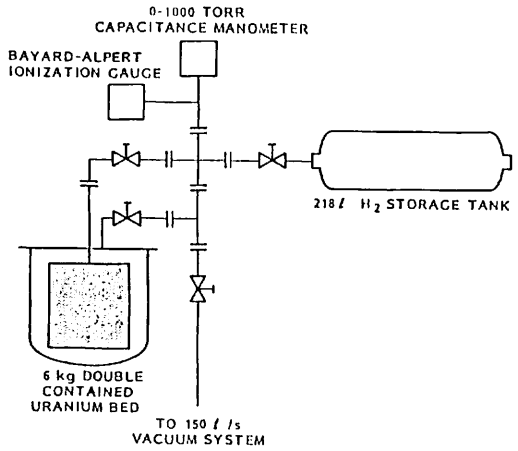

FIGURE 4: 6 kg URANIUM BED TEST LOOP

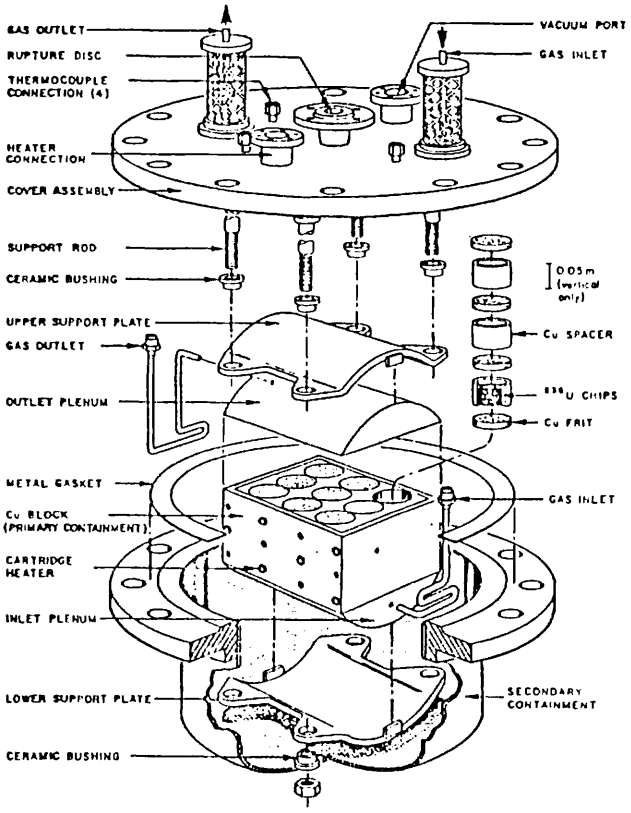

FIGURE 5: EXPANDED VIEW OF THE 6 kg URANIUM BED (4)

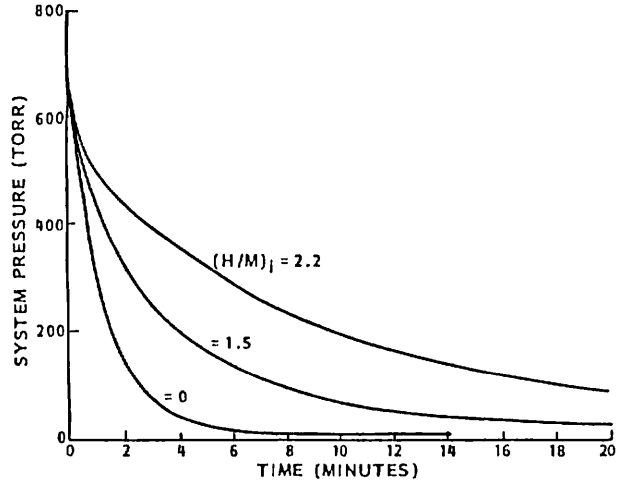

FIGURE 6: LOADING RATE DEPENDENCE ON INITIAL HYDROGEN/METAL $(H/M)_i$ RATIO ON 6 kg U BED (FILL PRESSURE 750 TORR; GAS FILLED SECONDARY CONTAINER)

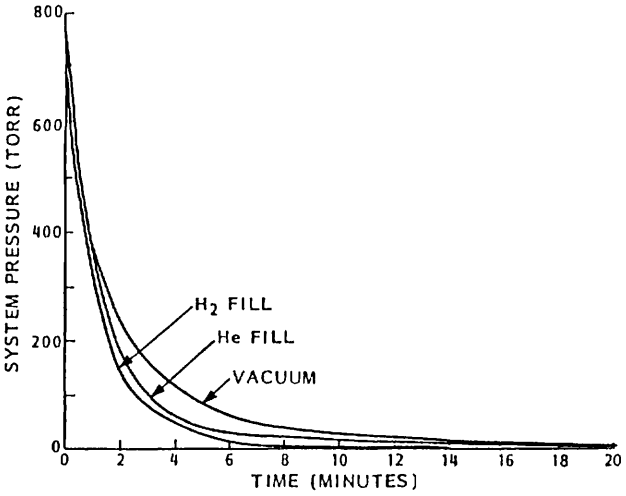

FIGURE 7: LOADING RATE DEPENDENCE ON 6 kg U BED SECONDARY CONDITION (FILL PRESSURE 750 TORR; INITIAL H/M = 0; FINAL H/M = 3/4)

The loading rate dependence on the initial hydrogen to metal ratio on the 6 kg LASL U bed is illustrated in Figure 6. The effect of the choice of gas in the secondary volume, or lack of it, during loading is shown in Figure 7; the concomitant change in temperature of the primary vessel is given in Figure 8. The higher thermal conductivity of hydrogen relative to helium results in a shorter charging period and a smaller increase in the primary vessel temperature. On the other hand, in the absence of a conducting fluid a significant reduction in the overall rate of loading and a corresponding increase in the primary vessel temperature are observed.

Heat transport again plays a major role in the rate of hydrogen regeneration. The critical factors are: intimate contact between heaters, the primary vessel wall, and the copper blocks containing the uranium powder; and, minimum thermal transport contact area between the primary and secondary containers. To recover hydrogen from the LASL bed required typically two days at a regeneration temperature of approximately 330°C. The recovery process was not carried out at a higher temperature due to the unavailability of a sufficient number of functional heaters on the primary container.

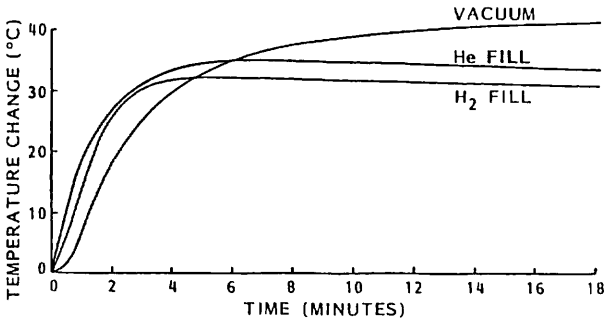

FIGURE 8: RELATIVE TEMPERATURE INCREASE IN 6 kg U BED PRIMARY CONTAINER FOR DIFFERENT CONDITIONS IN THE SECONDARY VOLUME

Two-Tier 660 g Bed

The two-tier, four-quadrant, 660 g uranium bed, designed, built and tested at Ontario Hydro, is shown in Figure 9.(5) The hydrogen loading behaviour of this bed was generally similar to the 15 g bed. The placement of a 5 μm filter between the tiers, in addition to the 5 μm filters at the inlet and outlet, resulted in restricting the flow and thereby limiting the overall rate of hydriding. Loading the uranium bed via both the inlet and outlet ports proceeds more rapidly. Essentially complete hydrogen recovery, from a fully loaded bed, at 375°C is achieved in 6 h.

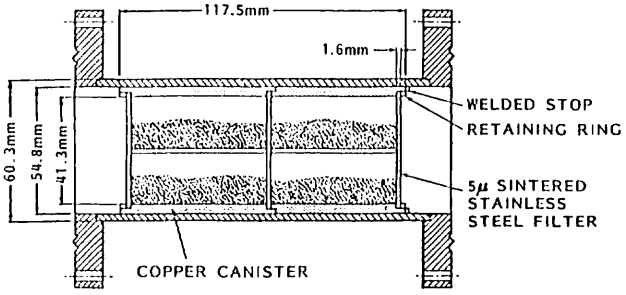

FIGURE 9: 660 g URANIUM BED

Simple 100 g Shipping Bed

The Amersham International Transportable 100 g uranium bed (6) was examined for its operating characteristics on a test loop similar to that shown in Figure 1. The uranium is contained within a stainless steel cylindrical vessel outfitted with a 70 mesh filter (~180 μm), a single inlet/outlet port, and a Cajon valve. The uranium bed, during transportation, is packed in a stainless steel, O-ring sealed, vacuum tight canister. The canister in

turn is placed amidst supportive, thick cork insulation within a galvanized steel shipping drum.

Loading the uranium bed to capacity in hydrogen to metal ratio increments of 0.78 requires approximately 15 min. A reduction in the rate of hydrogen loading in the presence of helium was similar to that observed with the 15 g bed. At a temperature greater than 350°C, the maximum rated storage capacity of the uranium bed (20 kCi equivalent hydrogen) is essentially recovered within 0.5 h.

TITANIUM GETTER BED

Titanium is a vigorous hydride former with a very low equilibrium pressure at ambient temperature. A dissociation pressure equal to atmospheric pressure is achieved at a temperature of approximately 650°C and hydrogen to metal ratio of unity.(7) Thus, titanium has been chosen as the primary or long-term storage medium at the TRF.

Simple 200 g Bed

A simple 200 g titanium bed, one quarter the scale of an Immobilization Tritium Container, was designed, constructed and tested.(5) The bed consisted of 200 g of titanium sponge contained within a stainless steel cylindrical chamber outfitted with 5 μm filters and an inlet and outlet port on either side of the vessel.

Hydrogen charging of titanium proceeded very rapidly. The system pressure dropped from an atmosphere to 100 Pa, with a hydrogen to metal increment upon loading of 0.25, within 100 s, largely independent of the bed temperature and composition (upto hydrogen to titanium ratio, H/Ti=1). Essentially complete hydrogen unloading, into the vacuum system, at approximately 550°C was achieved within 18 h (see Figure 10).

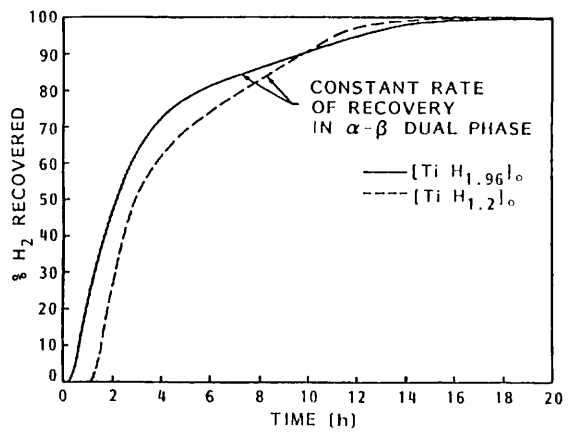

FIGURE 10: PERCENT H_2 OF INITIAL BED CONTENT RECOVERED DURING DISCHARGING OF 200 g TITANIUM SPONGE (TEMPERATURE ~500°C)

Numerous hydrogen transfers from the titanium bed to the 660 g uranium bed were conducted to simulate tritium transfer from a permanent storage medium to a temporary one. A direct, pump-less transfer (with initial H/Ti=1.2, regeneration temperature of 550°C, and an empty uranium bed at ambient temperature) required 18 h to transfer 83% of the hydrogen. Transfers using a Normetex circulation pump (circulating through both beds) required 19 h to transfer essentially all the hydrogen, in the absence and presence of helium.

DOUBLY CONTAINED URANIUM BEDS FOR TRITIUM SERVICE

A 500 kCi (18 500 TBq) uranium storage bed with secondary containment, shown in Figure 11, has been designed and built for tritium service at the TRF. At the time of writing the operating characteristics of the storage bed were being determined. The stainless steel primary vessel contains 3 kg of uranium partitioned within a two-tier, six-sextant copper block. The two-tiers are separated by a 180 μm stainless steel sintered porous filter. Loss of uranium particulates is controlled by the use of 5 μm filters staked onto the copper block. The uranium can be heated with the aid of band heaters placed on the primary vessel. The secondary vessel is made out of stainless steel outfitted with a port to permit evacuation of the secondary volume and penetrations for electrical and thermocouple feed-throughs. The primary vessel is surrounded by a number of dimpled stainless steel foils, which serve to minimize radiative heat loss during unloadings.

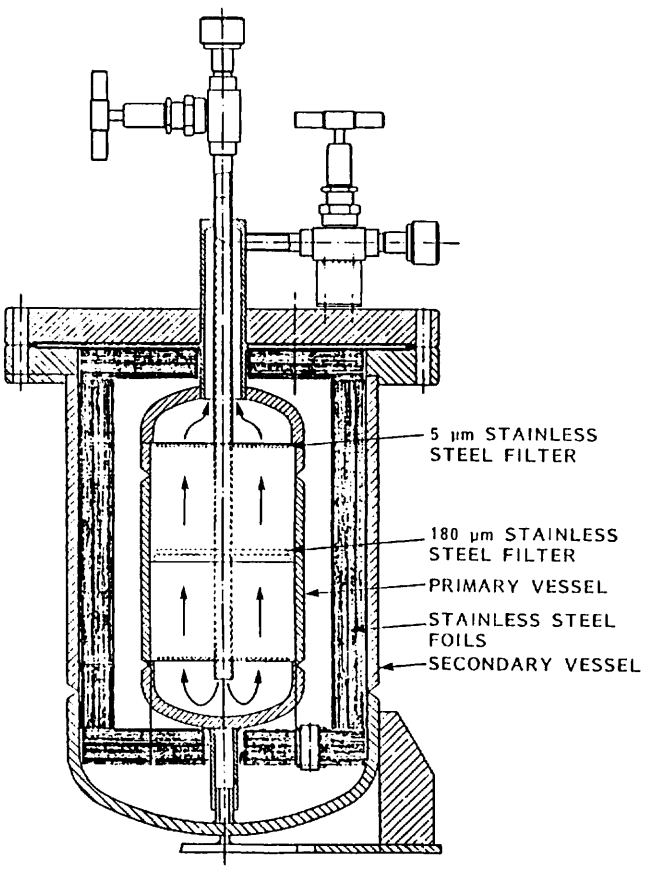

FIGURE 11: 500 kCi (18 500 TBq) DOUBLY CONTAINED URANIUM BED

A 5 kCi (185 TBq) doubly contained uranium bed, shown in Figure 12, has been designed for tritium service in the Ontario Hydro Research Division Tritium Laboratory. At the time of writing the bed, an entirely stainless steel construction, was in the

advanced stages of fabrication. The primary chamber contains 25 g of uranium with 5 μm sintered filters welded in place. The primary vessel is suspended from the weldments between the inlet and outlet ports and the secondary vessel. A band heater on the primary vessel will be used to heat the uranium. The secondary vessel is outfitted with penetrations for electrical and thermocouple feed-throughs and a flow port to permit evacuation of the secondary volume.

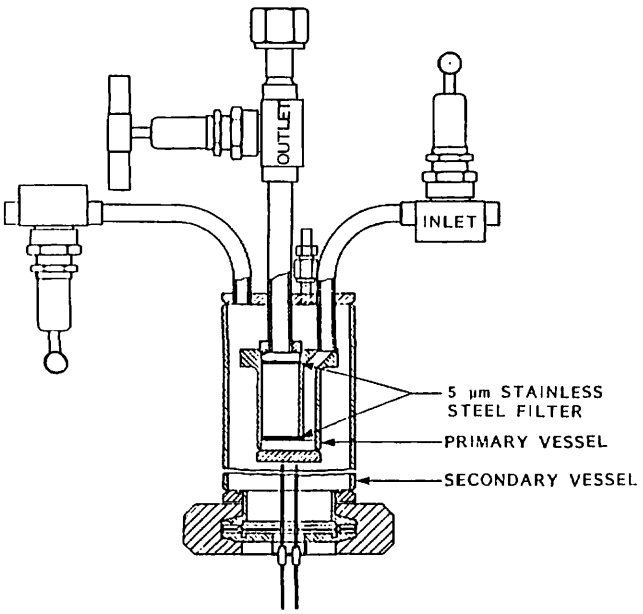

FIGURE 12: 5 kCi (185 TBq) DOUBLY CONTAINED URANIUM BED

CONCLUDING REMARKS

Uranium and titanium storage beds have good operating characteristics for application as temporary and permanent tritium storage media. Uranium, however, has certain drawbacks: it is a pyrophoric material, chemically toxic and therefore requires particulate control, and it is a controlled (nuclear) material. Currently work is underway to identify getter materials that are potential alternates to uranium with the following desired characteristics: dissociation pressure of 10 mPa and 0.1 MPa at 20°C and 400°C, respectively; negligible to modest expansion during hydriding, thereby reducing the pyrophoricity and particulate control concerns; good thermal conductivity; rapid hydriding and dehydriding kinetics; tolerant to impurities; stable in air; easy activation procedure; and minimum helium retention.

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