Asian Journal of Chemistry Vol. 22, No. 10 (2010), 7778-7786

Calibration of Experimental Model of Tritium Storage Bed

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During the water detritiation process most of the tritium inventory is transferred from water into the gaseous phase. Tritium is further enriched and finally extracted and safely stored. The control of tritium inventory is an important issue from several points of view, *viz.*, (i) tritium is an expensive material, (ii) safeguard-tritium is considered as nuclear material of strategic importance, (iii) safety-tritium is a radioactive material, (iv) requirements for documented safety analysis report (to ensure strict limits on the total tritium allowed) and (v) for evaluation of accident consequences associated with that inventory. Large amounts of tritium can be stored, in a very safety manner, as metal tritides. An experimental model of a tritium storage bed with integrated system for *in situ* tritium inventory accountancy was designed and manufactured at ICIT Rm. Valcea. The calibration curve and the detection limit for this experimental model of tritium storage bed were determined experimentally.

Key Words: Tritium, Calibration, "*In-situ*" accountability.

INTRODUCTION

A heavy water detritiation pilot plant is in commissioning/licensing stage at the Institute of cryogenics and isotopes technologies-ICIT Rm. Valcea (Romania) and the tritium inventory allowed by The Romanian Regulatory Body for this pilot plant is 2.2×10^{-3} g for commissioning stage and 6 g for operational stage.

Also, a project for construction of a tritium removal facility at Cernavoda NPP site was started in 2006, intending to be completed in the next 5 years. As storage materials the most used are depleted uranium (U-238), ZrCo, ZrNi and titanium (mostly used for long-term storage). A tritium storage bed must satisfy two important requirements (i) a high level of safety and (ii) an easy and reliable estimation of tritium content.

Tritium accountancy of the storage beds at tritium facilities is based either on removing of tritium followed by determination of pressure, volume, temperature and concentration (volumetric method) or on measuring the decay heat of tritium

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(0.324 W/g of tritium) using a constant heat flow calorimeter. The second is the most accurate accounting method available but the storage bed to be assayed must be removed from its process line and must be small enough to fit inside the calorimeter chamber.

An experimental model of a tritium storage bed with integrated system for *in situ* tritium inventory accountancy was designed and manufactured at ICIT Rm. Valcea. The calibration curve and the detection limit for this experimental model of tritium storage bed were determined experimentally. The experimental results are shown in Fig. 1.

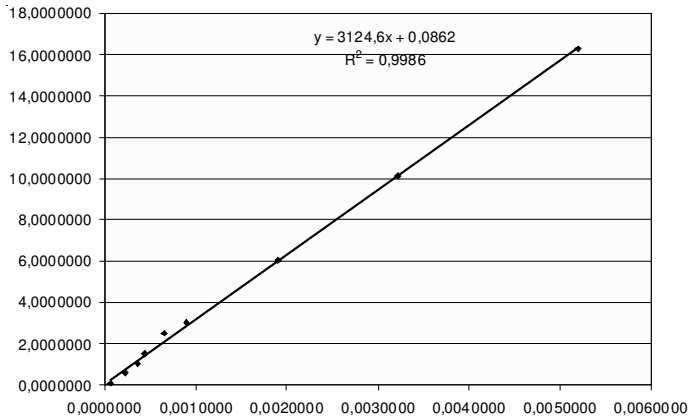

Fig. 1. Calibration curve of the experimental model of tritium storage bed

EXPERIMENTAL

Experimental model of tritium storage bed: The determination of the stored tritium inventory is made by measuring the heat generated inside the bed due to the β-decay of tritium (0.324 W/g of tritium). By its special design, the experimental model of tritium storage bed designed and manufactured at ICIT incorporates a system for measuring the temperature field inside the bed and also an internal electrical heat source to simulate the tritium decay heat for the calibrations purpose. This storage bed is suitable for performing "*in situ*" tritium accountability measurements.

A key requirement in the designing of the tritium storage bed is to achieve an uniform temperature distribution inside the bed. Thus, efficient heat dissipation is required. But storage materials disintegrate into a very fine powder upon hydriding process and powder is known to have a poor thermal conductivity. To overcome this disadvantage, the use of materials with high thermal conductivity is required. The optimum solution was to install a number of thin copper fins, equally spaced, between the two stainless steel wall cylinders of the storage vessel (Fig. 2). Uniform and thin layers of particles of titanium powder will be deposited on the surface of each of the thin Cu-fins.

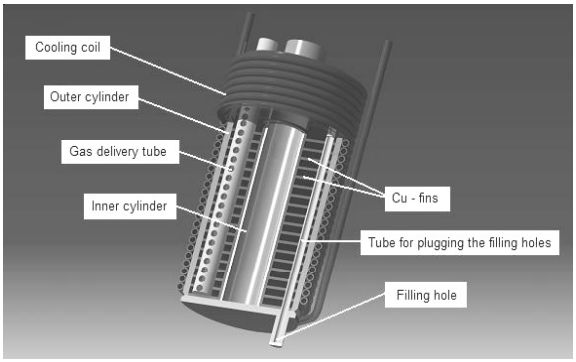

Fig. 2. Tritium storage bed

To remove the heat generated either by tritium decay or during the absorption of tritium, a gas cooling coil is wrapped around the outer wall of the primary containment.

An electrical heater is used to simulate the decay of tritium for calibration purposes. Several highly sensitive thermocouples were installed on the external surface of the inner cylinder and on the inner surface of the outer cylinder in order to monitor the temperature field inside the storage bed.

For optimum heat transfer, all components of the storage bed are brazed together. This experimental model of the storage bed was designed to assure a maximum storage capacity of 50 g of pure tritium.

The stored tritium inventory can be determined with different methods. The classical method involves the absorption of tritium in the storage bed as metal hydride. The extraction of tritium at high temperatures, recovered into a calibrated volume and then the temperature, pressure and concentration of tritium are measured. This method, even one of the simplest, has a lot of shortcomings *i.e.*, big effort, supplemental consumption of time and materials and the risk of tritium contamination. In addition, depending on the storage material, the temperature needed for the extraction of tritium can be very high, about 400 ºC in case of uranium, to about 700 ºC in case of titanium. Also, there is the possibility that not all the tritium can be extracted and a bigger risk is that of explosion, in case of loosing the tightness in the extraction installation.

RESULTS AND DISCUSSION

In present work, the developed tritium calorimeters which can determine the stored tritium inventory by measuring the heat emitted inside the bed as a result of β-disintegration of tritium (0.324 W/g of tritium). The measurement of the tritium inventory with this method is the most accurate method developed until now, with detection limits down to 0.003 g of tritium[1,2]. Although, there are some disadvantages, as follows: the storage bed has to be pulled-out of the circuitry of tritiated gas, the measurement can not be done on-line; the time of measurement is between

8 h and 7 days, depending on the quantity of tritium and the thermal properties of the storage bed and on its mass[1], which means the measurement is not made in real time; the bed has to have the geometric measures demanded by the measurement chamber of the calorimeter; high price of such calorimeter (between 200 and 300 US$).

Taking into account the disadvantages of the previous methods for measuring the tritium inventory, several laboratories in the world, handle and process tritium, developed different types of tritium storage beds provided with integrated systems for measuring the tritium inventory, which can determine the heat generated by the β-disintegration of tritium[1-3]. The systems for measuring the tritium inventory stocked in this type of storage beds are based on circulating a gas (usually helium) through a coil placed inside the bed and measuring the flow and the temperatures of the gas at its entry and at its exit (known as "in-bed gas flowing calorimetry"). The quantity of heat extracted with the cooling gas is measured as difference in gas enthalpy at its entry and exit. The calibration of this system is done with a heat source placed inside the bed, which simulates the heat emitted by the β-disintegration of tritium. The main advantages of this method are that the tritium measurement can be done on-line in real time and it is cost saving. Although, the measurement system by measuring gas and the systems for flow control and temperature measurement are quite complex. The determination of the calibration factor must be done in stationary conditions, which means that the gas flow must be done automatically in function of the storage bed temperature, so it remains constant. This involves precise measurement of the temperatures (at least three sensors), a controller for the adjustment functions implementation (hardware and software) and an execution element (with continuous adjustment). All these requests, added to the precision, calibration and certification requests for the measurement sensors, lead to a rise of price of such a tritium storage bed.

Another disadvantage of this method is the possibility of contamination with tritium of the circulating gas, by diffusion of tritium through the coil walls. In present tritium storage bed, we chose a method for the determination of the tritium inventory based on the measurement of the temperature field inside the bed. Besides, the constructive design of this storage bed allows the use of the "in-bed gas flowing calorimetry" method, by giving a double role to the cooling system of the bed. The main role of the system is to evacuate the heat generated by the absorption of tritium in the storage material. Obviously, to fulfill the role of measuring the heat generated by the β-disintegration of tritium, it has to be provided with the automated system for flow regulation, described above. The advantage of this experimental model of tritium storage bed over others similar models is the fact that the circulating gas is flowing on the outer side of the bed, away from the tritiated environment and reducing the risk of contamination of the circulating gas.

For both measuring methods which can be applied in this constructive design, *i.e.*, one based on the extraction of the heat with the cooling gas and the another

based on the measuring of the temperature field inside the storage bed. It is necessary to eliminate all the influences of the environment to the measurement. In order to achieve this, the storage bed has been insulated to reduce the loss of heat to a neglecting quantity. To minimize the loss of heat by radiation, the storage bed was covered with a glossy reflective foil used as a radiation shield.

We measured the temperature rise rates at the outer wall of the cylinder for several values of the injected power in the interest domain (0-16 W, corresponding to 0-50 g of tritium). The measurment was done graphically and represented the variation in time of the temperature of the storage bed on the domain of linear proportionality, from where we can determine the rising rates of the temperature T corresponding to every value of the power of the electric source. These values are equal with the slope of the line on the linear proportionality interval of the temperature in time. Here are the graphical representations of the temperature growth in time for several values of power injected in the storage bed: 0.1, 0.6, 1.0, 1.5, 2.0, 3.0, 6.0, 10.0 and 16.0 W (Figs. 3-11).

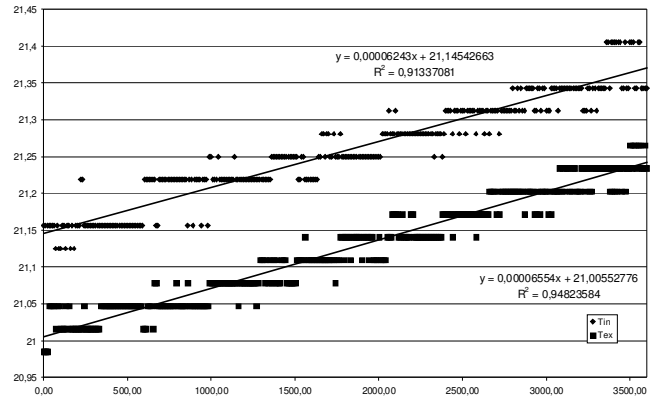

Fig. 3. Temperature *versus* time for P = 0.1 W

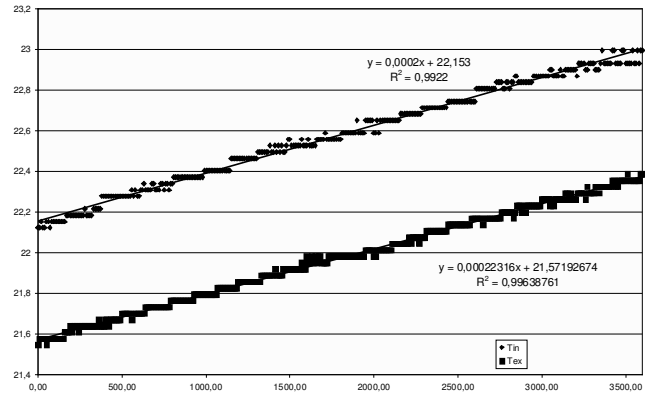

Fig. 4. Temperature *versus* time for P = 0.6 W

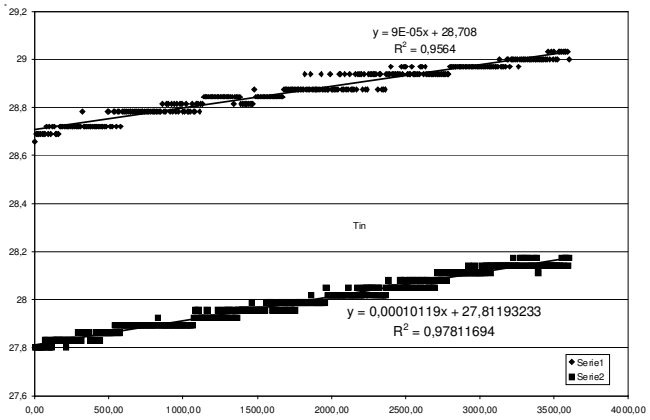

Fig. 5. Temperature *versus* time for P = 1.0 W

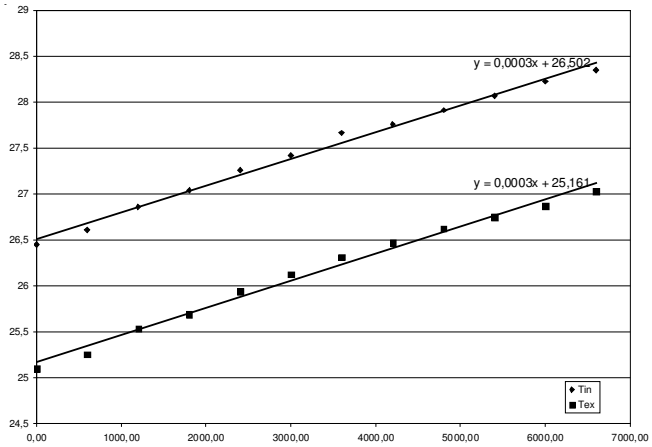

Fig. 6. Temperature *versus* time for P = 1.5 W

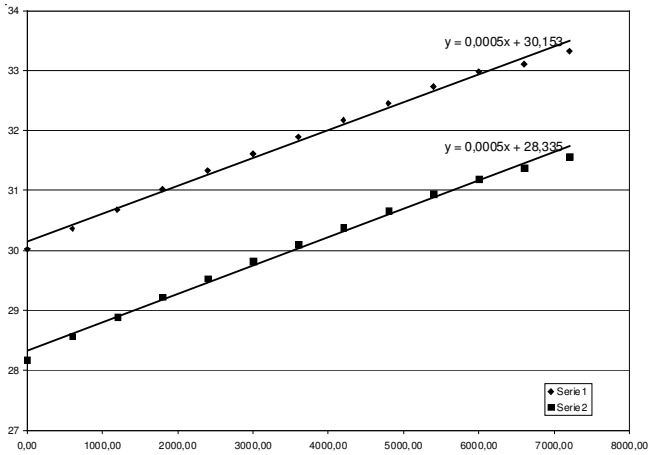

Fig. 7. Temperature *versus* time for P = 2.0 W

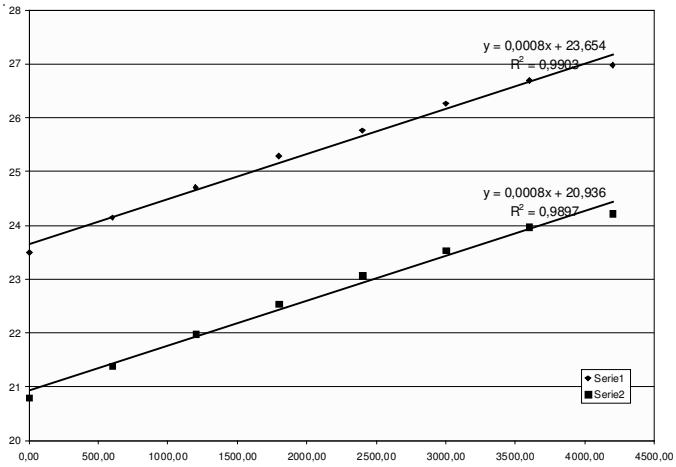

Fig. 8. Temperature *versus* time for P = 3.0 W

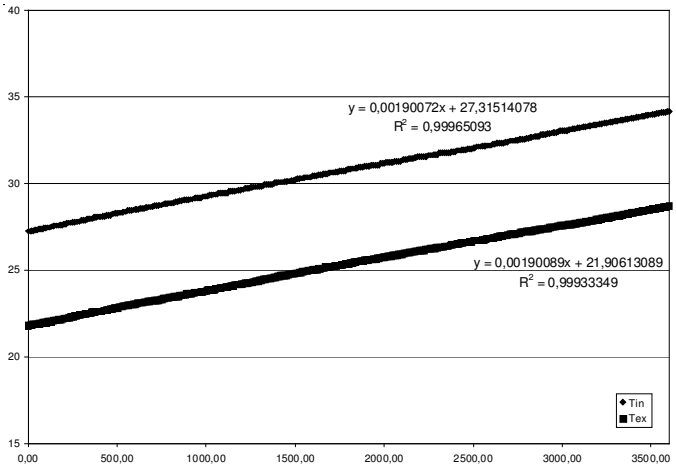

Fig. 9. Temperature *versus* time for P = 6.0 W

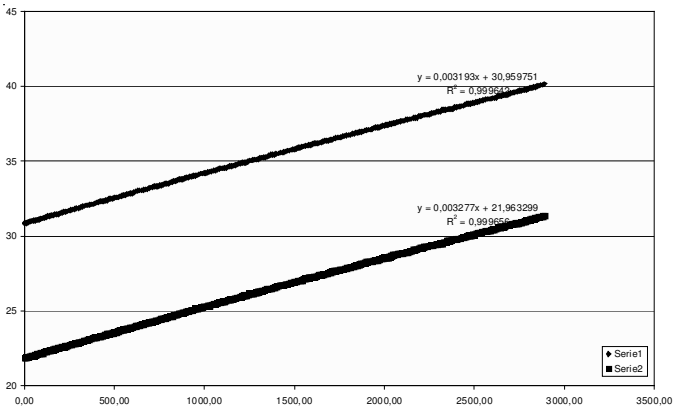

Fig. 10. Temperature *versus* time for P = 10.0 W

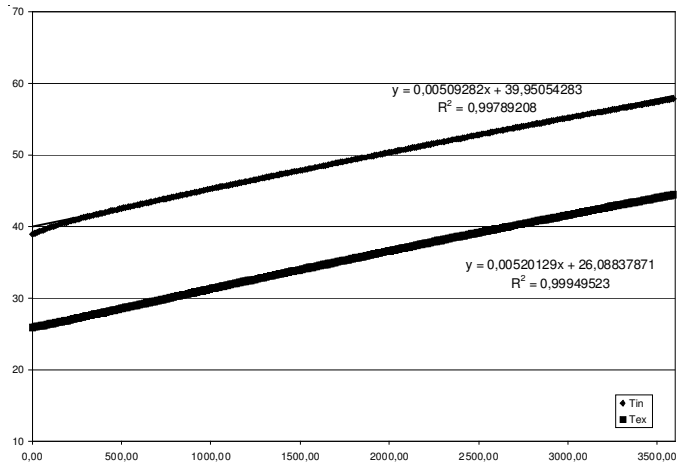

Fig. 11. Temperature *versus* time for P = 16.0 W

The experimental results are interesting because of the rise rates of the temperature corresponding to every value of the injected power. These are equal with the slope of the line on the linear domain of the variation in time of the temperature. We obtained the following graphic representation for the calibration curve of our experimental model of tritium storage bed:

This curve's equation is:

$$y = 3124.6x + 0.0862 \tag{1}$$

Compared with the formula:

$$W - W_{loses} = C\frac{dT_{ex}}{dt} \tag{2}$$

we obtain:

C[W·s/ºC]	3124.6
W_{loses} [W]	0.0862

The equation 2 is obtained from the case of a heat flow through a cylinder in transitory regime.

From now on, we can determine any value of the power generated by the heat sources uniformly distributed inside the storage bed and also the amount of stored tritium, by measuring the growth rate of the temperature on the outer wall, on a certain interval of time corresponding to the domain of linear proportionality.

$$W = 3124.6\cdot(dT/dt)_{Measured} + 0.0862 \tag{3}$$

We also estimated the detection limit for this experimental model of tritium storage bed, to *ca.* 0.7 g of pure tritium, which is a good result, compared with other models of storage beds.

An experimental model of a tritium storage bed with integrated system for *in situ* tritium inventory accountancy was designed and manufactured at ICIT Rm. Valcea. The storage capacity of this storage bed is 50 g of pure tritium. The calibration

curve and the detection limit for this experimental model of tritium storage bed were determined experimentally. The value obtained for the detection limit was about 0.7 g of tritium, which means 1.4 %. This method of calibration is simple one. It involves the use of 2-6 sensitive thermocouples, a digital voltmeter and a high accuracy ammeter and the results are more precise than when using the "in-bed gas flowing" calorimetric method.

Our result is consistent with other reported values of detection limits, situated between 1 and 3 % for tritium storage beds, but designed for in-bed gas flowing calorimetry.

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(*Received*: 18 February 2010; *Accepted*: 27 July 2010) AJC-8901