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Estimation of the Detection Limit of Tritium Storage Bed

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Large amounts of tritium can be stored, in a safely 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. An important issue is to know the exact amount of tritium stored inside the bed. The detection limit was determined by calculating the mean blank signal (zero heat injected in the electrical resistance) and the standard deviation of the blank signal and then treating the resulting data statistically.

Key Words: Tritium storage, Detection limit, "*in situ*" accountability.

INTRODUCTION

Tritium is a natural by-product of CANDU reactors and has a variety of commercial and industrial uses. Storage of tritium is an important issue, taking into account the facts that it is a radioactive material (and hazardous for people), but is also an element used in fusion experiments[1]. Tritium is obtained mostly from heavy water detritiation and until used, it must be stored safely. A safe and cost effective means for long term storage of tritium is needed. Storing tritium in a solid metal tritide is preferred to storing tritium as a gas, because a metal tritide can store tritium in a compact form and the stored tritium will not be released until heat is applied to increase its temperature to several hundred degrees centigrade. Storing tritium as a tritide is safer and more cost effective than as a gas. As storage materials the most used are depleted uranium (U-238), ZrCo, ZrNi and titanium (mostly used when long-term storage is envisaged)[2]. 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 or on measuring the decay heat of tritium (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[3].

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Estimation of detection limit using the calibration curve: The decay heat of tritium is determined by measuring the temperature increase of the thermal isolated storage bed, using the following formula:

$$W = \rho V c_p \frac{dT}{dt} \tag{1}$$

where W = power of the internal heat sources uniformly distributed inside the storage bed; ρ, V, c_p are the physical properties of the body of the storage bed (density, volume, specific heat, respectively).

Because the thermal isolation of the storage bed is not perfect, the power lost through the external surface of the bed (W_{lost}) must be introduced in the above equation:

$$W - W_{lost} = \rho V c_p \frac{dT}{dt} \tag{2}$$

or to be similar with the calibration curve equation,

$$\frac{dT}{dt} = \frac{1}{\rho V c_p} W - \frac{W_{lost}}{\rho V c_p} \tag{2'}$$

The terms W_{lost} and $1/(\rho V c_p)$ are experimentally determined by "injecting" precisely determined amounts of heat with the aid of calibration source[4,5]. So, the electrical heater is connected to an electrical power source and the input power can be adjusted by controlling the applied voltage. The calibration heat source is set to values between 0 and 16.2 W (corresponding to 50 g of tritium). For every value of delivered power, the temperatures inside the bed are measured for 1-2 h and the rise rate of this temperature was determined. The experimental set-up is shown in Fig. 1 and the experimental values obtained for the calibration of the tritium storage bed are presented in Table-1.

Fig. 1. Experimental setup for the calibration of the tritium storage bed

The values for temperature rise rate are plotted against the power delivered to the storage bed to determine the calibration curve. The graph of the calibration curve is shown in Fig. 2.

TABLE-1
EXPERIMENTAL VALUES OBTAINED FOR THE CALIBRATION OF THE TRITIUM STORAGE BED

Measurement No.	W_0 [watt]	dT_{ex}/dt [°C/s]
1	0.09943	0.00006554
2	0.60116	0.00022316
3	1.01037	0.00036785
4	1.51700	0.00044066
5	2.53010	0.00064885
6	3.03784	0.00089751
7	6.08440	0.00190089
8	8.10717	0.00268238
9	10.14158	0.00321808
10	16.26117	0.00520129

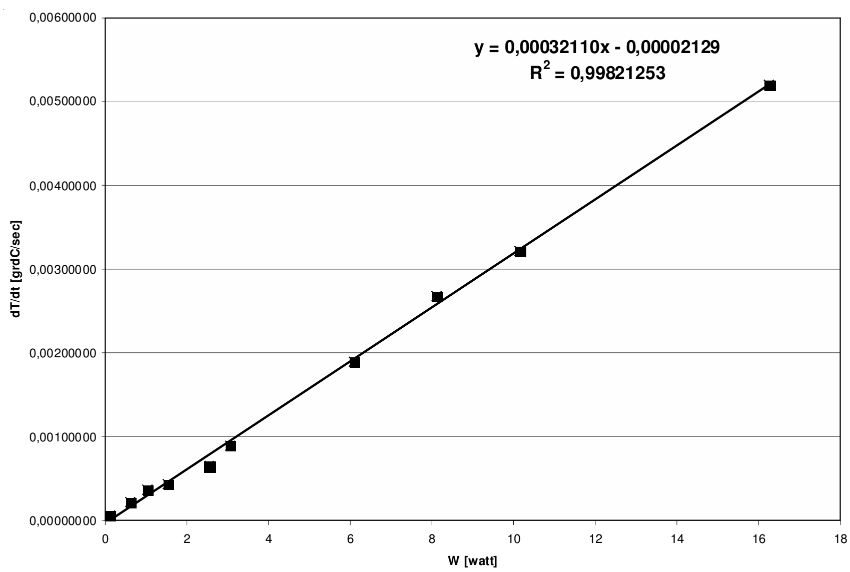

Fig. 2. Temperature rise rate *versus* the power delivered

Comparing the equation of the calibration curve with eqn. 2', it is suggested that y is the temperature rise rate dT/dt, x is the delivered power W, a is the slope of the calibration curve and constant b is the y intercept and is related to the inherent loses of heat, W_{lost}.

$$y = \frac{dT}{dt}$$

$$x = W$$

$$a = \frac{1}{\rho V c_p} = 0.00032110 \left[\frac{^\circ C/s}{W} \right]$$

$$b = \frac{W_{lost}}{\rho V c_p} = -0.00002129[^{\circ}C/s]$$

Linearity domain: Using calibration heat source, calibration curve y = ax + b were determined, where y = dT/dt is the measured signal (temperature rise rate), x = W is the known heat quantity delivered by the calibration power source. The linearity of the calibration curve was considered acceptable when the correlation factor R > 0.999.

Minimum detectable signal (y_m) is given by:

$$y_m = y_{blank} + k\sigma_{blank} \text{ (for detection with 99 \% confidence: k = 3)} \quad (3)$$

To determine mean blank signal, y_{blank} and standard deviation of blank signal, σ_{blank}, we performed 10 temperature measurements without any power delivered, over extended periods of time and the temperature rise rate was determined for each measurement (Table-2). In the following graph (Fig. 3) is presented an example of noise signal measured:

TABLE-2
VALUES OF THE BLANK SIGNAL (TEMPERATURE RISE RATE) FOR THE 10 MEASUREMENTS, THE VALUE OF MEAN BLANK SIGNAL AND THE VALUE FOR THE STANDARD DEVIATION

Measurement No.	Blank signal (ºC/s)	Mean blank signal (ºC/s)	Standard deviation (ºC/s)
1	9E-06	4.10E-06	4.202E-06
2	-3E-06		
3	-2E-06		
4	8E-06		
5	3E-06		
6	5E-06		
7	2E-06		
8	5E-06		
9	5E-06		
10	9E-06		

Treating the resulting data statistically, the mean blank signal (y_{blank} = 4.10E-06 ºC/s) was obtained and the standard deviation of blank signal (σ_{blank} = 4.202E-06 ºC/s). The value obtained for the minimum detectable signal from eqn. 3 is: y_m = 1.67E-05 ºC/s.

The detection limit (DL) is the lowest quantity which can be measured with reasonable statistical certainty. Using the slope (a) from calibration curve and taking into account the heat loses (W_{lost} = -b/a), we can estimate the detection limit from the following equation:

$$DL = \frac{y_m - y_{blank}}{a} + W_{lost} = k\frac{\sigma_{blank}}{a} + W_{lost} \quad (4)$$

With the values obtained previously, we estimate the detection limit to 0.222 W or 0.685 g of pure tritium.

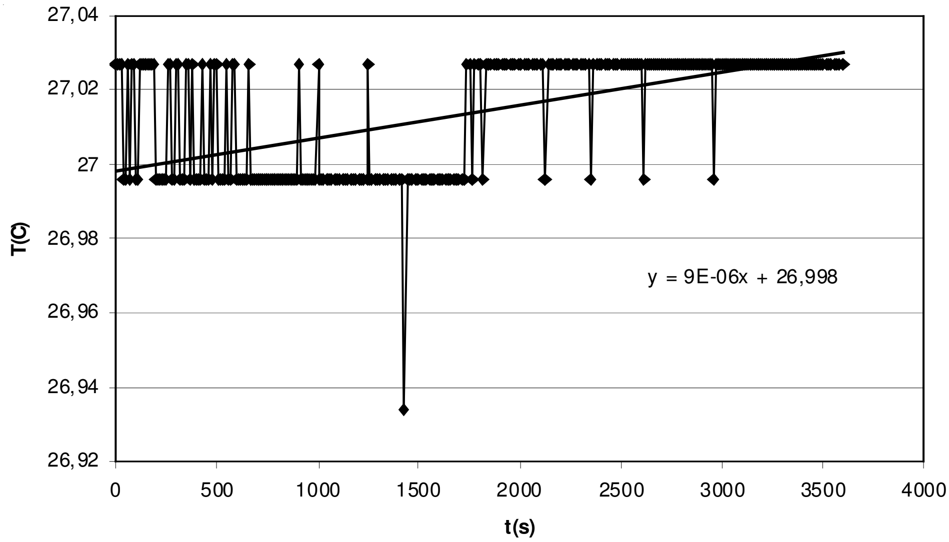

Fig. 3. Storage bed temperature *versus* time without delivering heat

Conclusion

An experimental model of a tritium storage bed with integrated system for *in situ* tritium inventory accountancy was designed and manufactured at ICIT Ramnicu Valcea. The storage capacity of this storage bed is 50 g of pure tritium. The detection limit for this experimental model of tritium storage bed was determined experimentally. The value obtained for the detection limit was about 0.7 g of tritium, which means 1.4 %. This model of tritium storage bed can be used in the Experimental Pilot Plant for separation of tritium and deuterium from ICIT Ramnicu Valcea and also at others Tritium Removal Facilities, as the one that is in commissioning stage at Cernavoda Nuclear Power Plant.

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