# IMPROVEMENT OF TRITIUM ACCOUNTANCY TECHNOLOGY FOR THE ITER FUEL CYCLE SAFETY ENHANCEMENT

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#### Abstract

In order to improve the safe handling and control of tritium for ITER fuel cycle, effective "in-situ" tritium accounting methods have been developed at Tritium Process Laboratory in Japan Atomic Energy Research Institute under one of the ITER-EDA R&D Tasks. A remote and multi-location analysis of process gases by an application of laser Raman spectroscopy developed and tested could provide a measurement of hydrogen isotope gases with a detection limit of 0.3 kPa for 120 seconds analytical periods. An "in-situ" tritium inventory measurement by application of a "self assaying" storage bed with 25 g tritium capacity could provide a measurement with a required detection limit less than 1 % and a design proof of a bed with 100 g tritium capacity.

# 1. INTRODUCTION

In the ITER design, the first attempt to build a complete fusion fuel cycle, which consists of processes which are fueling, burning (by fusion reaction), pumping, purification (with recovery from tritiated impurities), isotope separation and storage, was made. From the viewpoint of operation and safety, tritium accountancy control will be required for the design [1,2].

Tritium-rich hydrogen isotope gas will be moving continuously with a fairly large flow rate (200 Pa·m³/s maximum) through the systems of the ITER fuel cycle under the plasma operation. On the other hand, tritium inventory limits will be set for the individual (or some specified) components with operational and/or regulatory purposes. A method for dynamic measurement of fuel process gas with a short analytical period will be required to control fuel process components. When the method is also expected to be used for tritium inventory control even during plasma operations, a feature of uninterrupted measurement will be required. Also a static measurement of tritium with a good accuracy will be necessary for a periodical tritium tracking of the whole fuel cycle loop including the vacuum vessel components. To simplify the tritium accountancy process and estimate accurately the amount of tritium retained and/or solved in the components in a reasonable time and with practical processes, it will be required that the storage system has a feature of "self-assaying" tritium.

There were two projects held at the Tritium Process Laboratory (TPL) in Japan Atomic Energy Research Institute to establish the technological base for such tritium tracking and process system control under ITER-EDA. One was to develop a remote and multi-location analysis system of hydrogen isotopes and tritiated molecular impurities in process gases by application of laser Raman spectroscopy [3]. The other was to develop "in-situ" tritium inventory measurement by application of a "self assaying" storage bed [4, 5]. Here, the brief specifications and performance of the developed systems are reported.

# 2. DESCRIPTION AND PERFORMANCE OF DEVELOPED SYSTEMS

### 2.1 Remote and Multi-Location Process Gas Analysis System

A Remote and Multi-location Process Gas Analysis System of using laser Raman spectroscopy has been developed at TPL/JAERI to monitor and/or control fuel process components. It has a feature that gas analysis can be carried out simultaneously at plural analytical points with one system. Comparing current conventional methods for qualitative and quantitative analysis of process gas, such as gas chromatography, mass spectrometry, this system has advantages of much shorter analytical period and no requirement of additional systems for sampling, evacuating and exhaust gas processing.

Also the detector can detect almost all light signals from the sample gas flowing in the pipe continuously, i.e., dynamic inventory control of process components is possible with this system [3].

Laser Raman spectroscopy using optical fibers and a multi-channel detection system equipped with a liquid nitrogen cooled CCD is applied to this analytical system to enable remote and multi-location analysis of hydrogen isotope gases in a process gas pipe inside a glove box. Figure 1 show the optics module to be set in a analytical point with a cell developed for this system. The cell attached to the process pipe has four sapphire windows, for transfer of laser beam and Raman scattering light, which can stand differential pressure of 2 MPa. The Raman scattering light from each analytical point is transferred through a fiber array of six step index fibers (core/clad diameter  $114/125~\mu m$ , length:20 m) into a monochrometer. The Raman scattering lights from the four different analytical points are dispersed by a grating (1200 grooves/mm) and reflected on separated areas of CCD chip as schematically shown in figure 2 (up).

The first remote and multi-location analysis system was installed in the Isotope Separation System (ISS) of TPL. Tests for monitoring gas composition were carried out by using two optics modules set on the gas lines from top and bottom of the ISS distillation columns. Pressure and temperature of gases in the cells were in ranges of 10 - 100 kPa, 283 - 303 K, respectively. Acquisition time was set between 60 - 300 sec. The Raman spectra of different analytical points, which were transferred to the different CCD areas, were shown in figure 2 (down). The spectra were well distinguished from each other without any cross-talk peaks. From the noise level at a range where no Raman peak exists, the estimated detection limits was, about 0.3 kPa for  $H_2$ , HD and  $D_2$  with an acquisition for 120 sec, which was equivalent to a concentration of about 0.3% for a process gas with an atmospheric pressure.

Figure 3 shows the analytical results of the ISS product gas stream. Gas composition of ISS product gas stream was successfully measured with every 2 minutes interval and without extraction of any sample gases. The time dependence of gas composition fractions obtained from the spectra data above agreed well with the results of simulation analysis and the feasibility of this remote and multi-location analysis system was demonstrated

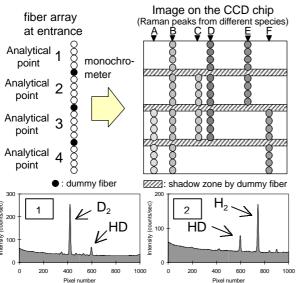


Figure 2. Schematic images of spectra on CCD detector (up) and spectra measured at analytical points 1 and 2 (down) of ISS.

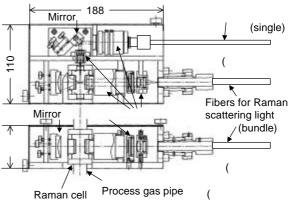


Figure 1. The optics module and a Raman cell for the remote and multi-location analysis

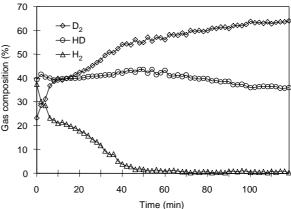


Figure 3. Measured trend of hydrogen isotope composition of the outlet gas from the bottom of the ISS column by the remote and multi-location analysis system.

# 2.2 Tritium Storage Bed for "In-bed" Gas Flowing Calorimetry

A "self assaying" storage bed was developed. This system has been developed to establish a safe and simple tritium accounting method for tritium storage system. "In-bed" gas flowing calorimetry was applied to a scaled ZrCo bed with 25 g tritium capacity [4, 5]. In the principle design of this calorimetric system, helium gas is circulated through a pipe inserted inside the primary vessel containing zirconium-cobalt tritide to remove decay heat of tritium. The tritium inventory is evaluated by the temperature difference at a steady state between inlet and outlet of the circulated He gas. Comparing volumetric method (Pressure, Volume, Temperature and Concentration) commonly used for tritium accountancy of the storage beds at tritium facilities in the world [7-10], this tritium accountancy method has advantages in the following points; 1) simple operation without requirement to move tritium from the system, 2) measurement of the whole amount of tritium stored in the bed at once and 3) continuous removal of tritium decay heat to avoid overheating the ZrCo that results in increase the gas pressure inside. It has been demonstrated using the scaled ZrCo bed that measurement within an accuracy of ±1 gram against 100 grams tritium storage for an ITER design requirement could be carried out with this method [4, 5].

Based on the results of the demonstration tests, a ZrCo bed of "In-bed" gas flowing calorimetry, which has a normal tritium storage capacity of 100 grams as ZrCoT<sub>1.5</sub> ( max.  $\sim 200 \text{ g T}_2 \text{ as ZrCoT}_3$  ,  $\sim 35$ mol of hydrogen isotopes gases), was designed for ITER, as shown in figure 4. The primary vessel contains about 3500 g of ZrCo powder and about 1500 g of copper balls (1 mm in diameter) for improvement of thermal conductivity. Other structure was the same as the 25 g T<sub>2</sub> storage bed tested.

Figure 5 shows the flow diagram of the whole "In-bed" gas flowing calorimetry system for tritium inventory measurement. Though, the He flow rate for the demonstration tests was 4 std. l/min, about 20 std. 1/min will be required for operation of the designed 100 g bed because of larger decay heat. In addition, a vacuum pump will be connected at the outlet of the process gas from the beds to maintain its hydrogenation / dehydrogenation performance. Because disproportionation of ZrCo possibly takes place under high hydrogen pressure and high temperature [11], e.g. when regenerating of the bed. To avoid the dispropotionation, the pressure at the outlet has to be kept low by using this vacuum pump.

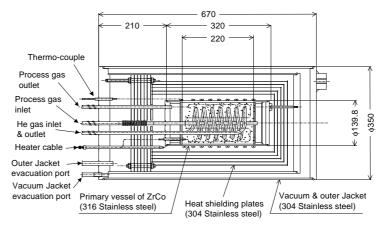


Figure 4. The cross section view of the designed ITER tritium storage bed. (unit: mm)

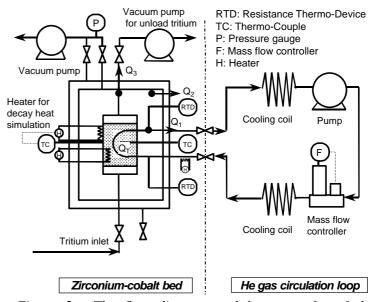
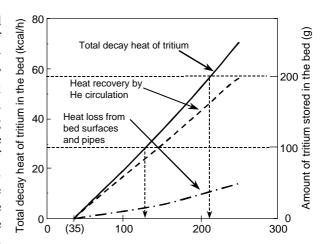


Figure 5. The flow diagram and heat transfer of the designed 100 g storage bed with "in-bed" calorimetric system. ( $Q_T$ : decay heat of tritium,  $Q_1$ : heat removed by He flow,  $Q_2$ : heat transferred by radiation from the surface,  $Q_3$ : heat transferred through piping.. Calibration was carried out with the heater for decay heat simulation.)

Figure 6 shows the relationship evaluated by analysis between the total decay heat of tritium stored and the temperature of the center part of the ZrCo stored section in the primary vessel under a calorimetric measurement operation using some empirical formulas obtained from the experimental data of 25 g storage bed. The heat is removed by both He circulation and radiation/thermal conduction of metal parts. As shown in figure 6, in this system, radiation and heat transfer through the structural materials would give rather little correspondence to the accountancy performance because more than 80 % of decay heat is removed by He gas circulation. The inlet temperature of He gas is designed to be kept at constant temperature of 35 °C with a small heater to avoid the effect of fluctuation of room temperature. Then the result shows that even when 200 g (maximum) of pure tritium is loaded into the bed the ZrCo (tritide) temperature in the



Temperature of the center part of ZrCo in the bed (°C) Figure 6. The evaluated relationship between the total decay heat of tritium stored and the temperature of the ZrCo stored section under a calorimetric measurement operation with a circulated He flow of 20 std. l/min (the outer and vacuum jackets are evacuated).

primary vessel will become less than 220 °C. This temperature is low enough to keep the hydrogen equilibrium pressure inside the ZrCo bed far below the atmospheric pressure and also high enough to give a proof of accountancy measurement with an accuracy less than 1% at the 100 g storage.

# 3. CONCLUSIONS

Two projects were successfully completed at TPL/JAERI to establish the technological base for such tritium tracking and process system control under ITER-EDA. Successful development of the remote and multi-location analysis system with an application of laser Raman spectroscopy would make it possible to provided qualitative and quantitative analysis of process gas with a good accuracy, a much shorter analytical period and no requirement of additional systems for sampling, evacuating and exhaust gas processing. Successful development of the "in-situ" tritium inventory measurement system with an application of a "self assaying" storage bed would make it possible to provide simple operation for accountancy of the whole amount of tritium stored in the bed without moving tritium from the system and to design of the ZrCo bed with a 100 g tritium capacity with a proof of sufficient tritium decay heat removal and a good accuracy.

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