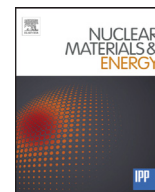




Contents lists available at ScienceDirect

Nuclear Materials and Energy

journal homepage: www.elsevier.com/locate/nme

Development of quantitative analysis method for tritium using by hydrophobic platinum catalyst

Yuki Edao*, Yasunori Iwai, Takumi Hayashi

Tritium Technology Group, Directorates of Fusion Research and Development, Japan Atomic Energy Agency, Tokai, Ibaraki, 319-1195, Japan

ARTICLE INFO

Article history:
Available online xxx

Keywords:
Tritium
Hydrophobic catalyst
Quantitative measurement
Accuracy

ABSTRACT

Various methods of tritium measurement have been applied depending on a chemical form of tritium. A method combined oxidation catalyst and water bubblers has been used as one of the most quantitative analysis methods for gaseous tritium. The major technical shortcoming with the conventional methods is the lowering of quantitative accuracy caused by adsorption of produced tritiated vapor on hydrophilic oxidation catalyst. Then, we developed an organic-based hydrophobic platinum catalyst. The catalyst enhanced its hydrophobicity to prevent absorption of water vapor on catalyst surface. In order to ensure to oxidize tritium, we additionally devised 1) the particle size of the catalyst made small to be around 1 μm and 2) intentional addition of hydrogen. As the result of experimental verification of the proposed method, we succeeded to achieve the high accuracy of more than 99.9% in the quantitative measurement of tritium. The developed tritium measurement system is applicable widely for various tritium measurements, estimation of tritium generation rate at TBM, tritium behavior in various materials and so on.

© 2016 The Authors. Published by Elsevier Ltd.

This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

Various methods of tritium measurement are applied corresponding to the concentration, the chemical form, the state and environment. Gaseous tritium measurement includes a method to measure ionization current generated by beta rays of tritium such as ionization chamber and proportional counters and also a method to measure radioactivity of a liquid sample collected tritium by liquid scintillation counting. A liquid scintillation counter is superior in measuring low concentration of tritium present in environment [1–3]. Then, a method combined oxidation catalyst and a trap such as an adsorbent, a cold trap or a water bubbler is used as one of the most quantitative analysis methods for tritium [1–4]. The method is not suitable for real-time monitoring but for measurement of tritium in a wide range of concentrations. The method also enables to distinguish the chemical forms between gaseous tritium and tritiated water vapor [2, 3]. In the method tritium is oxidized by oxidation catalyst. Then oxidized tritium is collected in a water bubbler and tritium collected in bubblers is analyzed by liquid scintillation counting. Copper oxide and hydrophilic platinum catalyst are generally used as tritium oxidation catalyst in a conventional method [4–6]. However, the major technical short-

coming with the conventional methods is the lowering of quantitative accuracy caused by adsorption of produced tritiated vapor on oxidation catalyst having hydrophilic property. The adsorption leads to incorrect mass balance of tritium due to adsorption of oxidized tritium on oxidation catalyst and caused misunderstanding of tritium behavior due to the delayed desorption of oxidized tritium from oxidation catalyst [6]. In order to respond the strong need of tritium measurement with high accuracy to understand the transient behavior of tritium released from tritium breeding material, the absorption of produced tritiated vapor on oxidation catalyst should be avoided as much as technically possible.

For this purpose, we developed an organic-based hydrophobic platinum catalyst to be applied for quantitative tritium measurement system. In the present study, we devised a high quantitative tritium measurement system and demonstrated the performance of the measurement system.

2. Experimental

2.1. Devisal for quantitative tritium measurement system

A quantitative tritium measurement system was proposed. It composed of a catalytic reactor packed with hydrophobic platinum catalyst and two water bubblers connected in series. In the catalytic reactor, tritium in air was completely oxidized to be tritiated

* Corresponding author.

E-mail address: edao.yuki@jaea.go.jp, edao.yuki@qst.go.jp (Y. Edao).<http://dx.doi.org/10.1016/j.nme.2016.06.013>2352-1791/© 2016 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

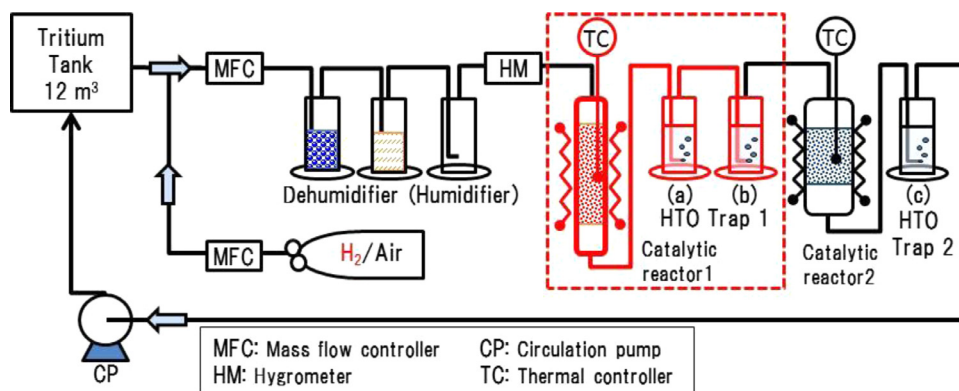


Fig. 1. Experimental apparatus to determine the performance of tritium oxidation. MFC: mass flow controller; HM: hygrometer; TC: temperature controller and CP: circulation pump.

water vapor (HTO). The water bubblers acted as the selective trap of HTO from air. In order to avoid retention of oxidized tritium by adsorption, we developed an organic-based hydrophobic platinum catalyst. The catalyst enhanced its hydrophobicity to prevent absorption of oxidized tritium on catalyst surface. In addition, the hydrophobic platinum catalyst can maintain the activity in the presence of water vapor due to its hydrophobicity, whereas it is well known that hydrophilic platinum catalyst considerably lowers the activity due to the covering of active sites by water molecules. In order to oxidize tritium completely in the reactor, we carefully selected the particle size of the catalyst. This is because an increase in contact surface area per packed volume enhances the efficiency of reaction in a case the diffusion in pores is the rate controlling step. It has been well demonstrated the efficiency of reaction has a dependency on hydrogen concentration. Smaller hydrogen concentration causes lower efficiency of reaction. The concentration of tritium to be oxidized is generally below the natural hydrogen content in air. In order to enhance the efficiency of reaction and to downscale a catalytic reactor, initial addition of hydrogen was proposed to oxidize tritium efficiently.

2.2. Performance test

Fig. 1 schematically illustrates the catalytic reactor flow system to demonstrate the performance of the proposed quantitative tritium measurement system. In the catalytic reactor flow system, the quantitative tritium measurement system we proposed is boxed by a dashed line. The catalytic reactor flow system has two catalytic reactors. The first catalytic reactor is the core of the quantitative tritium measurement system and the second catalytic reactor is to oxidize the tritium unreacted in the first catalytic reactor. The catalyst packed in the first reactor was a hydrophobic platinum catalyst which is supporting Pt directly on styrene divinylbenzene copolymer (Pt/SDBC). The catalyst used in the tests was commercially named Kogel catalyst, test-manufactured by SHOKO Co., Ltd., Japan [7]. The Pt/SDBC catalyst was 1.0 wt% Pt/SDB particles with the diameter of 1 mm ϕ . In order to uniform temperature distribution in the catalytic reactor by heating from the outside surface of the reactor, the catalytic reactor was a slender shape with the inner diameter and length of 15.8 mm and 500 mm, respectively. The volume of the Pt/SDBC catalyst packed in the catalytic reactor was 94 cm³ to oxidize tritium completely. In the second reactor, Pt/Al₂O₃ catalyst was packed to oxidize HT unreacted in the first reactor completely. The Pt/Al₂O₃ catalyst was 0.5 wt% Pt/Al₂O₃ pellets manufactured by N. E. Chemcat Corporation, Japan. The inner diameter and length of the second catalytic reactor were 56.5 mm and 150 mm, respectively. The volume of the Pt/Al₂O₃ catalyst packed in the catalytic reactor was 200 cm³. Both

catalytic reactors were thermally insulated. The temperature of catalyst layer was controlled by a temperature controller. The temperature of catalyst layer was monitored with a thermocouple. Careful attention was paid to the low thermal stability of Pt/SDBC catalyst due to polymeric substrate. The maximum temperature of Pt/SDBC catalyst layer was 100 °C. The temperature of Pt/Al₂O₃ catalyst layer was kept at 200 °C. The Pt/Al₂O₃ catalyst layer was continually washed out by wet air from HTO trap 1, then tritium adsorption on the surface of Pt/Al₂O₃ catalyst was kept to be negligibly small.

A mixture gas of tritium balanced in air was supplied from a tank of 12 m³ to the catalytic reactor flow system. The system was capable of flow rates of 100–1500 cm³/min controlled with a mass flow controller (8500MC, KOFLOC, Japan). The concentration of the tritium in air was approximately 370 Bq/cm³. The concentration of water vapor in a mixture gas supplied was controlled with/without a humidifier (a wet/dry condition) after water vapor was removed by two dehumidifiers packed with silica gel beads and molecular sieves 5A pellets, respectively. The hydrogen was intentionally added for the hydrogen concentration of the mixture gas to be a desired value (0–5000 ppm). The mixture gas passed through the first catalytic reactor packed with Pt/SDBC catalyst. Gaseous tritium (HT) converted to the oxide by the Pt/SDBC catalyst and collected by the water bubblers (a) and (b) of the HTO Trap 1 as shown in Fig. 1. Gaseous tritium unreacted in the first catalytic reactor fed into the second reactor packed with Pt/Al₂O₃ catalyst and was completely oxidized by the Pt/Al₂O₃ catalyst, the oxidized tritium was collected by the water bubbler (c) of the HTO Trap 2. The tritiated water sample was taken from the HTO Trap 1 and Trap 2 by pipetting. The weight of the sample was measured taking variations in the amount of tritiated water by pipetting into consideration. The tritium activity in the HTO Trap 1 and Trap 2 was measured by a liquid scintillation counter (LSC6100, Aloka Co., Ltd., Japan). The water bubbler used to enhance efficiency of tritiated vapor trapping was a gas washing bottle that could contain a volume of 500 ml with a cylindrical glass filter with the pore size of 100–160 mm manufactured by SIBATA SCIENTIFIC TECHNOLOGY LTD. When the bubbler was filled with water of 200 ml at room temperature, the height to water surface from bottom of the filter was 37 mm.

In order to establish the quantitative tritium measurement system, tritium should be completely oxidized in the catalytic reactor and produced tritiated vapor should be completely trapped in the HTO traps. In evaluation of accuracy of proposed quantitative tritium measurement system, the system consists of two subsystems which are catalytic reactor packed with hydrophobic catalyst and water bubblers connected in series. Hence, the accuracy of proposed quantitative tritium measurement system can be assessed

Download English Version:

<https://daneshyari.com/en/article/7987627>

Download Persian Version:

<https://daneshyari.com/article/7987627>

[Daneshyari.com](https://daneshyari.com)