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Results of experimental study on detritiation of atmosphere in large space

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Abstract

In order to obtain data on tritium removal from the atmosphere of a room, which is needed for designing a reliable effective atmosphere detritiation system in a fusion reactor and for detailed analysis on its safety, intentional tritium release experiments have been carried out in a controlled space called Caisson under various atmosphere conditions at the Tritium Process Laboratory (TPL) in Japan Atomic Energy Research Institute (JAERI). In case there is no tritiated water vapor in the released tritium gas, tritium was ideally removed by constant ventilation in spite of atmospheric conditions and residence time. On the other hand, when tritiated water vapor existed in the released tritium, residual contamination on the wall of the Caisson was detected and it was found that it depended on the initial humidity in the Caisson. This tritium removal behavior was successfully analyzed by considering the adsorption and desorption reaction rates of tritiated water on the wall by the constant ventilation.

Keywords: Fusion safety; Tritium; Confinement; Tritiated water; Detritiation; Adsorption; Desorption

1. Introduction

In future fusion reactors of high safety and acceptability, safe confinement of tritium is one of the key issues for realizing a fusion reactor, and tritium should be well controlled so that it is not released excessively into the atmosphere and to prevent workers from excess exposure. Tritium will be handled under a multiple confinement system in a fusion reactor and each level of confinement will have its own detritiation system [1]. This concept of the multiple confinement has been successfully adopted in tritium facilities in the world [2,3]. The Tritium Process Laboratory (TPL) of the Japan Atomic Energy Research Institute (JAERI) is a facility licensed to handle 22.2 PBq of tritium, which has also a multiple confinement system [4], and it has been accumulating the safety experiences for more than 12 years without any accidental tritium release to rooms and the environment since its foundation.

A room and/or a building will be important as a final confinement barrier of tritium to the environment, but only a few reports have been presented on actual tritium behavior in such a large

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confinement space [5]. In order to establish a more effective and reasonable tritium safe-handling and confinement system, actual data of tritium behavior in a large confinement space should be accumulated systematically with various ventilation conditions and atmospheric conditions, and a reliable computer analysis code for tritium behavior in a large space is needed to be developed based on the accumulated data.

We carried out the intentional tritium release experiments in a radiologically controlled area of 3000 m^3 of the Tritium System Test Assembly (TSTA) in Los Alamos National Laboratory (LANL) under the US–Japan collaboration program [6], from which we could learn about the actual tritium behavior in the room. But there exists some difficulties in understanding the results of the experiment well because they were carried out under only a few limited conditions.

In order to understand the tritium behavior in a large space clearly, a series of systematic investigations on tritium behavior in the well-controlled medium-scale simple space were found to be needed to link the results of basic research on tritium behavior to its behavior in the actual room. Therefore, the Caisson Assembly for Tritium Safety study (CATS) was planned and installed in TPL/JAERI in 1998, which consists of the Caisson, a 12 m^3 leak-tight box for the investigation of tritium behavior. The main purposes of the experiment with CATS are the accumulation of data on tritium behavior from initial diffusion to final removal under various atmospheric conditions to establish its analysis code and the performance of the equipments for tritium confinement and removal. We have carried out the intentional tritium release under the simple N_2 atmosphere [7] and the analytical study using the improved three-dimensional eddy flow model [8]. As a result, tritium behavior in the simple atmosphere could be completely analyzed [7–9].

As the next step, the experiment under typical Japanese atmospheric conditions such as high humidity is in progress.

In this paper, tritium removal behavior in the Caisson under various atmosphere conditions is discussed comparing the experimental and analysis results. The effect of the tritiated water fraction in the total released tritium is also discussed.

2. Experimental

2.1. Apparatus

The CATS consists of the 12 m^3 Caisson and a neighboring glove box (GB), which is installed in a leak-tight room of the TPL. Fig. 1 shows the conceptual layout of the CATS. The Caisson is a 2.6^{W} m $\times$ 2.0^{D} m $\times$ 2.3^{H} m stainless steel leak-tight box and the GB is of dimension 2.0^{W} m $\times$ 0.95^{D} m $\times$ 1.8^{H} m. The Caisson is connected to a ventilation system and a conventional detritiation system, with which the ventilation condition of 12–50 m^3/h can be produced. The Caisson is controlled by a negative pressure of approximately 30 Pa to atmospheric pressure in the room. A humidity control system is also connected to the Caisson to produce typical Japanese conditions. The amount of tritium released in the Caisson in each experiment is permitted to 37 GBq. Seven nude type ion chambers (TM0–6 in Fig. 1) and a normal gas flow type ion chamber (GM) are installed inside and outside the wall of the Caisson to measure the tritium concentration. Furthermore, to measure the tritiated water vapor concentration in the Caisson, a bubbler system connected to the Caisson with five bubblers is used. Each bubbler captures tritiated water vapor by flowing a sampling gas of 100 $cm^3\ min^{-1}$ and the system can automatically change the bubbler every 30 min.

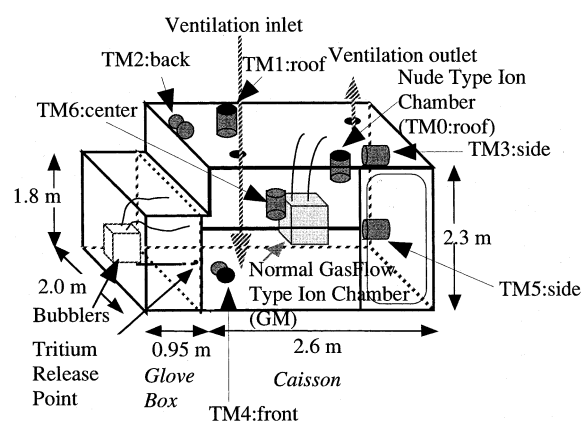

Fig. 1. Structure of the CATS and location of tritium monitors and tritium release point.

Table 1
Tritium release experimental conditions

Chemical form	T_2+N_2 (with tritiated water ~3.0%)
Amount of released tritium	2.6 GBq
Atmospheric condition	Dry air, humid air (~600 ppm)
Residence time	2.5 h to 1 week
Detritiation system	50 m^3/h (circulation)

2.2. Experimental conditions

In the present series of experiments, a 4 cm^3 stainless steel container was prepared and 2.6 GBq of tritium diluted by N_2 was filled in it. In every experiment, 2.6 GBq of tritium in the container with N_2 was released into the Caisson ventilated to 50 m^3/h by purging with N_2 gas of 150 $cm^3\,min^{-1}$ for 1 min. In the case where there is no tritiated water in the released total tritium, a cold trap containing a copper ball was installed in the tritium release system. Therefore, when tritium was released into the Caisson, tritiated water was removed by the cold trap. The local total tritium concentration is measured by TM0–6 and GM, and the tritiated water vapor concentration is measured by the bubbler system. Table 1 shows the experimental conditions of intentional tritium release.

3. Results and discussion

The present series of intentional tritium release experiments in the Caisson was carried out under a humidity condition of 1–600 ppm. Tritium (2.6 GBq) diluted by N_2 was released in every experiment with or without tritiated water vapor of around 260 MBq. After leaving the released tritium in the Caisson for some time (from several hours to 1 week), it was removed by a conventional detritiation system every time. From the present experiments, the humidity and tritiated water vapor dependency on tritium behavior could be obtained as discussed below.

3.1. Tritium removal behavior without tritiated water vapor in released tritium gas

Fig. 2 shows the typical tritium removal behavior trace obtained with the conventional detritiation system of 50 m^3/h. TM, GM and ideal trace represent the indication of the nude type ion chamber, the normal gas flow type ion chamber and the ideal tritium removal curve, respectively. In the case where there is no tritiated water vapor in the released tritium gas, both in dry air and in humid air atmosphere, the released tritium into the Caisson was removed ideally by 50 m^3/h of a conventional detritiation system within 180 min as shown in Fig. 2. Residual tritium contamination on the wall of the Caisson was not detected. Dependence of humidity in the atmosphere of the Caisson on this removal behavior was not observed so far as no tritiated water vapor was released. Therefore, the released tritium gas did not cause any adsorption and desorption on the wall of the Caisson. The experiment as a function of the residence time of tritium gas in the Caisson was also carried out to measure the conversion rate of tritium gas to tritiated water and to know the effect of the produced tritiated water on the detritiation. The released tritium gas was left in the Caisson from several hours to a maximum of 1 week. Fig. 3 shows the result after 1 week. Even after 1 week, the released tritium gas was removed ideally by the detritiation system. Though the conversion rate of tritium gas to tritiated water

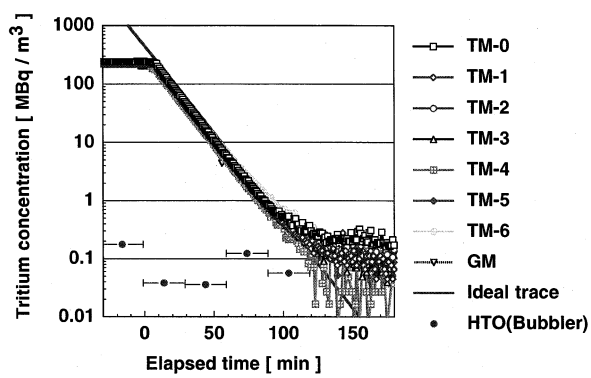

Fig. 2. Typical tritium removal behavior in the dry air atmosphere (humidity: 12 ppm) by 50 m^3/h of detritiation.

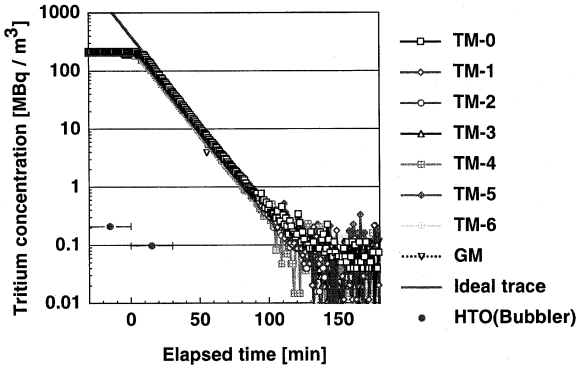

Fig. 3. Tritium removal behavior in the dry air atmosphere (humidity: 10 ppm) by 50 m^3/h of detritiation (residence time: 1 week).

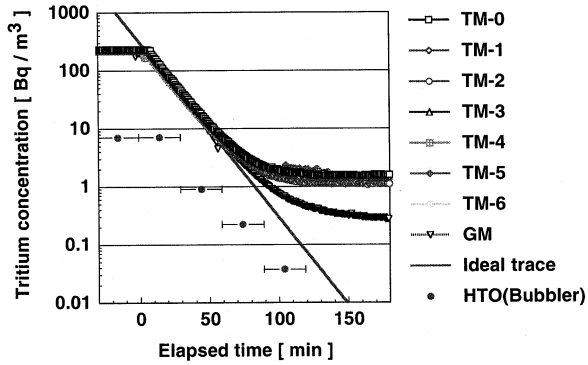

Fig. 5. Tritium removal behavior in the humid air atmosphere (humidity: 582 ppm) by 50 m^3/h of detritiation (HTO in the released tritium: 3.0%).

was evaluated to be less than 0.01% per day in the Caisson, the effect of the produced tritiated water to tritium removal behavior was not detected remarkably.

3.2. Tritium removal behavior with tritiated water vapor in released tritium gas

When tritiated water vapor existed in the released tritium, as shown in Figs. 4 and 5, tritium removal from the Caisson took a longer time than the ideal one and residual tritium contamination on the stainless steel wall of the Caisson was detected. Adsorption and desorption between tritiated water vapor and the wall is considered to cause the increase of the removal time, which is called the soaking effect. Initial humidity of the atmosphere in the Caisson was found to affect this effect. When the initial humidity was very low, such as less than 1 ppm, tritium in the Caisson was removed much slowly than the case without any tritiated water vapor in the released tritium as shown in Fig. 4 in comparison with Fig. 2 or 3. When the initial humidity in the Caisson was high, quicker removal could be done in comparison with the low initial humidity case, which might be because tritiated water vapor was diluted by the humidity and tritiated water vapor became more difficult to adsorb on the wall of the Caisson than in the dry atmosphere case. A large difference between TM and GM in this tritiated water vapor release experiment was observed. This is considered to have appeared due to the difference of memory effect between TM and GM, GM has much lower memory effect than TM because GM has a pump for purging its ion chamber while TM does not. Therefore, TM measured the surface contamination of the ion chamber with atmospheric tritium concentration. It means that, at least 100 min after detritiation as shown in Fig. 4, the indication of TM represents mainly the wall contamination by tritiated water.

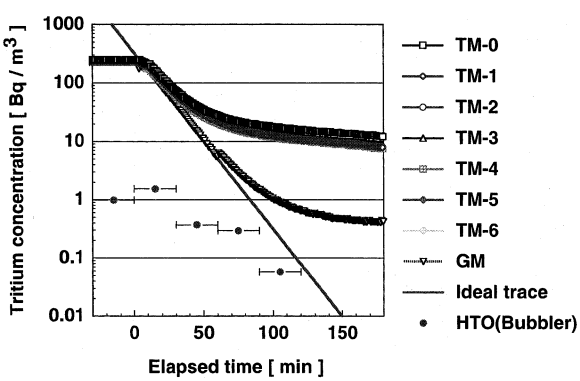

Fig. 4. Tritium removal behavior in the dry air atmosphere (humidity: 1 ppm) by 50 m^3/h of detritiation (HTO in the released tritium: 0.4–1.0%).

3.3. Analytical approach

The primary tritium removal behavior was investigated by a simple simulation code [9]. In an ideal case for tritium removal, tritium concentration is decreased exponentially as given by Eq. (1),

$$\frac{dW_i}{dt} = -W_i \frac{F}{V} \quad (1)$$

where V, F, W and t represent the volume of the confinement, the flow rate of the ventilation, the amount of tritium and the ventilated time, respectively. In the practical case, the removal tritium was slowed by several factors. From the results in the Caisson, in the case without any tritiated water vapor, the tritium removal behavior was not affected by the actual low conversion rate of tritium gas to tritiated water and by the soaking effect of tritium gas on the wall of the Caisson. However, in the case of tritiated water vapor existing, the soaking effect of tritiated water on the wall of the Caisson affected the tritium removal behavior. Therefore, we considered the adsorption and desorption reaction of tritiated water on the wall of SUS304, applied Freundlich's equation as given by Eq. (2),

$$\frac{dW_j}{dt} = -W_j \frac{F}{V} - k_a A \frac{W_j}{\sum_{j=1}^{3} W_j} \left(\sum_{j=1}^{3} C_j \right)^{0.53} + k_d q_j \quad (2)$$

where W_j, A, k_a, k_d, C_j and q_j represent the amount of water vapor component (H_2O, HTO, T_2O), the surface area, the adsorption reaction rate, the desorption reaction rate, the water vapor concentration (H_2O, HTO, T_2O) and the amount of the adsorbed water on the wall of the Caisson, respectively. Analyzing the tritium gas removal by Eq. (1), and analyzing the tritiated water vapor removal by Eq. (2), traces shown in Figs. 6 and 7 were achieved, in which the experimental results are also shown. The analyzed tritium removal behavior was in good agreement with the experimental results by considering the soaking effect under constant ventilation in various humidity atmospheres.

4. Conclusions

Systematic intentional tritium release experiments were planned and performed with the Caisson, a 12 m^3 leak-tight box, to investigate the tritium removal behavior under constant ventilation in various conditions.

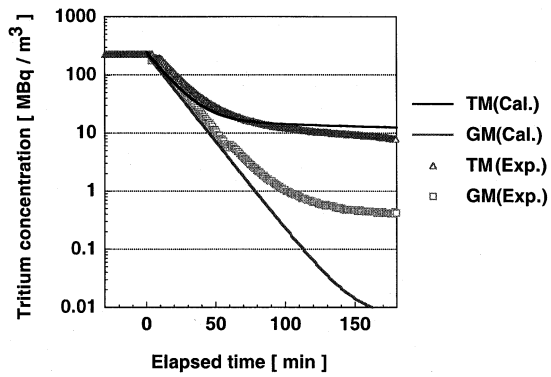

Fig. 6. Analytical results of tritium removal behavior in the dry air atmosphere (humidity: 1 ppm) compared with experimental data (HTO in the released tritium: 0.4–1.0%, k_a: 2.26×10^{-9} ms^{-1}, k_d: 2.68×10^{-5} s^{-1}).

1. In the case there is no tritiated water vapor in the released tritium, tritium was removed ideally from atmosphere of the Caisson by the detritiation system with 50 m^3/h. Humidity and residence times of tritium in the Caisson were found not to have an affect on the tritium removal behavior in this case.
2. When tritiated water vapor existed in the released tritium, it took a much longer time to remove tritium from the Caisson compared with the case where no tritiated water vapor is released. In this case, it was found that the humidity in the Caisson affected the tritium removal behavior.

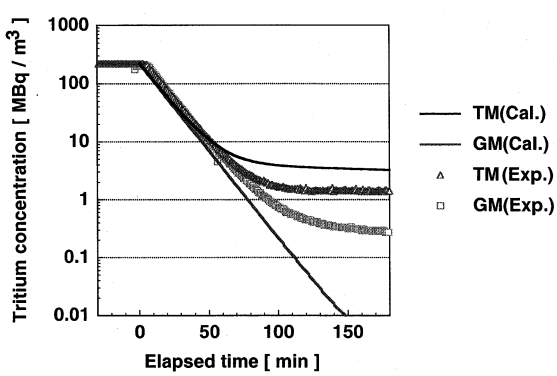

Fig. 7. Analytical results of tritium removal behavior in the humid air atmosphere (humidity: 582 ppm) compared with experimental data (HTO in the released tritium: 3.0%, k_a: 2.26×10^{-9} ms^{-1}, k_d: 2.68×10^{-5} s^{-1}).

3. By considering the adsorption and desorption reactions of tritiated vapor on the wall, the effect of humidity when tritiated water vapor was released together with tritium gas was clearly understood from the analysis.

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