Estimation of tritium inventory in exhaust detritiation system for fusion test device in the initial tritium recovery operation

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As one of the radiation safety issues at nuclear fusion facilities, the development of the tritium handling technique is indispensable. A small amount of tritium is produced by the deuterium plasma experiment in a large fusion test device and is exhausted from the vacuum vessel. From the viewpoint of radiation safety and public acceptance, tritium in the exhaust gas could be recovered by the exhaust detritiation system (EDS). Most of the tritium recovered by EDS is stored in the wastewater tanks as tritiated water. Then they are fractionated into the wastewater containers for delivery to the Japan Radioisotope Association once a year. However, some of the tritiated water would have remained in EDS. The residual tritium would be the issue of the dismantlement of EDS in the future. Thus, the amount of residual tritium in EDS was estimated in the initial tritium recovery operation. Since most of the wastewater containing tritium remains in the storage tanks, the residual wastewater was the main tritium inventory. Also, the tritium in the molecular sieve used in EDS was estimated to be less than 0.1 GBq. Thus, the decontamination of molecular sieves would be preferred when dismantling the EDS.

Keywords; exhaust detritiation system, fusion test device, tritium removal, tritiated water, tritium contamination, tritium inventory

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1. Introduction

To understand the high-temperature plasma physics, nuclear fusion plasma research using hydrogen isotope gases has been conducted in fusion research facilities all over the world. As one of the large fusion test devices, the Large Helical Device (LHD), which has the largest helical type superconducting magnet coil, started the deuterium plasma experiment on March 7, 2017 [1]. In the deuterium plasma experiment by LHD, tritium is produced by the D-D fusion reaction and is exhausted from the vacuum vessel. The tritium is the radioactive material and then decays into helium-3 with a half-life of 4500 days [2]. Thus, from the viewpoints of radiation management and public acceptance, exhausted tritium must be removed by the tritium removal system [3]. The tritium removal system has been in operation in some tritium handling facilities and fusion test facilities. The tritium removal technology is oxidation by catalysts and water absorption by absorbents such as molecular sieves [4, 5, 6]. As the tritium removal system for the LHD deuterium plasma experiment, the exhaust detritiation system (EDS) [7] has been installed and operated since 2016 before the start of the deuterium plasm experiment. The EDS mainly consists of MS (molecular sieves) type and PM (polymer membrane) type as the tritium removal system. The former system uses molecular sieves for tritium removal and is used during the plasma experiment. The latter system uses a polymer membrane for tritium recovery and is used during the vacuum vessel maintenance activity after the plasma experiment. Both systems recover the tritium as the chemical form of water. Most of the tritium recovered by both MS and PM type systems is collected in the temporary storage tanks. Then they are fractionated from the tanks into the wastewater container. The wastewater containers are delivered to the Japan Radioisotope Association (JRIA) [8] once a year. However, a part of the tritium has remained in the EDS. The contamination and inventory of tritium in EDS would be the issues of the dismantlement in the future. Thus, we estimated the tritium inventory in EDS during the initial operation phase.

2. Tritium removal system and tritium measurements

2.1. Exhaust detritiation system [7]

The EDS for LHD has been designed by JGC Corporation. The process flow diagram of the exhaust detritiation system is shown in Fig. 1. The exhaust fan system in Fig. 1 blows the process gas through the EDS as well as the ventilation air in the facility into the stack.

The MS system mainly consists of a low-temperature (473 K) oxidation reactor, a high-temperature (723 K) oxidation reactor, two absorbent columns for switching packed with molecular sieves 5A (416 kg), a gas storage system, and a molecular sieves regeneration system. The low-temperature oxidation reactor converts hydrogen gas into water vapor whereas the high-temperature oxidation reactor converts hydrocarbons into water vapor [7]. In the MS system, as the hydrogen concentration becomes higher, the temperature of the oxidation reactor rises due to the combustion heat of the hydrogen. Since the combustion process of hydrogen in the high-temperature oxidation reactor would cause problems of overheating and heat removal, a low-temperature oxidation reactor for hydrogen was installed in the first stage. The process

flow rate is up to 20 Nm^3/h.

The PM system consists of a low-temperature (473 K) oxidation catalyst, a blower, a two-stage screw type compressor, a cold condenser, 20 hollow fiber type polymer membrane modules (UM-C10, length: 1116 mm, diameter: 90 mm, UBE Industry Ltd.). The process gas is wet air and the tritium chemical form is water vapor. There are no hydrogen gas and hydrocarbons in the ventilation from the vacuum vessel [7]. However, hydrogen gas which was not used in plasma experiments is occasionally released into the PM system. Thus, a low-temperature oxidation catalyst was installed. The PM system handles the ventilation gas from the vacuum vessel containing tritium released during maintenance of the LHD and NBIs (Neutral Bean Injection systems). The process flow rate is up to 300 Nm^3/h. The detritiation factor of EDS has been achieved to be more than 20, which is the requirement specification [9, 10]. The detail of EDS is described elsewhere in ref [7].

As a common utility system in EDS, a wastewater delivery and storage system is equipped. It consists of three temporary receiver tanks, a large storage tank, a wastewater measuring tank, and some liquid transfer pumps as shown in Fig. 2. To monitor the variation of wastewater amount in these tanks, a certain quantity of water was placed in these tanks before starting the deuterium plasma experiment. The water that regenerated the molecular sieve in the MS system, the condensed water from the heat exchanger, and the recovered water from the polymer membrane system were collected in each temporary receiver tank. When the wastewater amount in the temporary receiver tanks reaches the upper limit, the liquid transfer pump is automatically activated and transfers the wastewater into the large storage tank. For the delivery of the wastewater to JRIA, the wastewater of 0.05 m^3 in the large storage tank is manually transferred to the wastewater measuring tank and is divided into two 25-liter wastewater containers. After the tritium concentration in wastewater is measured, these containers are delivered to the JRIA.

2.2. Tritium monitoring and measuring instruments

To evaluate the tritium amount in EDS, the water bubbler systems are installed in EDS. The original water bubbler systems [11] are installed at the inlet of the MS system and the outlet of the MS and PM systems. MARC 7000 (SDEC, France) is installed at the inlet of the PM system. These water bubbler systems were operated for one day to two weeks depending on the LHD operation or the schedule of the LHD maintenance activity. The volume of sampling gas was controlled between 0.5 ~ 1 m^3. The sample water volumes for the original water bubbler system and MARC 7000 were approximately 20 g and 100 g per one bubbler column, respectively. The sample water used deionized water. After a certain period of collection operation, sample water and scintillation solution (UltimaGold LLT, Perkin Elmer) were mixed in a ratio of 1:1 in a 20 cm^3 polyethylene vial. Then the cocktail sample was measured by a liquid scintillation counter (Tri-Carb 4910TR, Perkin Elmer). The background water for the liquid scintillation measurement is the same as the sample water of the water bubbler system. The counting time for tritium measurement was totally 50 minutes. The same procedure was

applied for the measurement of tritium activity in wastewater. The detection limit of the water sample was less than 10^{-2} MBq/m^3-water. Then the detection limit of tritium concentration was on the order of 1 Bq/m^3-gas.

3. Results and discussion

The deuterium plasma experiment due to LHD started on March 7, 2017. Then, produced tritium was exhausted from the vacuum vessel of LHD [12]. After the end of the plasma experiment period, the vacuum vessel was opened for maintenance from September 2017. Before the workers enter the vacuum vessel, the vacuum vessel was ventilated by room air to reduce the tritium concentration to below 2 kBq/m^3. During the ventilation, tritium was continuously released from the vacuum vessel walls [13]. Released tritium was recovered by EDS. The recovered tritiated water was fractionated into the wastewater container on January 16, 17, and August 13, 14, 2018. In this study, the period of operation from March 7, 2017, to August 14, 2018, is covered as the initial tritium recovery operation.

3.1. The tritium recovery and inventory in the MS and the PM systems

As mentioned above, tritium decays with a half-life of 4500 days. Therefore, the tritium amount decreases with time. The tritium inventory in EDS considering tritium decay would be expressed by the following equation:

$$A_n = A_{n-1} \times \left(\frac{1}{2}\right)^{\frac{a}{4500}} + E_n^{in} - E_n^{out} . \quad (1)$$

Here, A_n is the tritium inventory in the n-th week, E_n^{in} is the received tritium amount at the inlet of EDS in the n-th week, E_n^{out} is the tritium amount through EDS in the n-th week, a is the sampling period, normally 7 days. The variation of tritium inventory in the MS and the PM systems is shown in Fig. 3. The main measurement errors may arise from counting errors in the liquid scintillation counter and the accuracy of the flow measurement. The error in the amount of tritium estimated for each measurement was evaluated to be less than 1 MBq from the standard deviation of the counts measured by the liquid scintillation counter. The error may be much less than 1% of the estimated tritium amount. The accuracy of the flow measurement device is 2%. The error bars in the figure are hidden by the symbols and cannot be almost viewed. In these data, tritium fractionation operation does not include. The data of the received tritium amount at the inlet of the MS and the PM systems are used from the previous report [12, 13]. The tritium inventory in the MS system increased during the period of the plasma experiment because the tritium recovery operation was carried out. The MS system was shut down after the plasma experiment and the tritium inventory in the MS system was gradually decreased owing to the tritium decay process. The tritium recovery operation due to the PM system has been carried out since September 2017. Then, the tritium inventory in the PM system increased following the end of the plasma experiment. Since the tritium amount released from

the LHD vacuum vessel during the maintenance was much lower than that during the plasma experiment, the tritium inventory in the PM system was maintained at a low level in comparison with the MS system. The amount of tritium inventory in EDS, which is calculated from the difference between the tritium amount received into and released from EDS, was estimated to be approximately 2.40 GBq. The decay of tritium in the EDS during the evaluation period, 1.5 years, was estimated to be approximately 0.147 GBq. Then, the tritium inventory in the MS and the PM systems was estimated to be approximately 2.04 GBq and 0.207 GBq, respectively, at the end of the LHD maintenance activity. The tritium was recovered as the tritiated water into the temporary receiver tanks. Then they were delivered to the large storage tank when the water level in the tanks reached the upper limit. Thus, it is difficult to estimate the tritium concentration in each temporary receiver tank because of the repeated collection and transfer of tritiated water with different tritium concentrations.

3.2. The tritium concentration in the wastewater

The average tritium concentration and the amount of fractionated wastewater are summarized in Table 1. The standard deviation of the tritium concentration for each measurement was less than 10 MBq/m^3. Two fractionation operations were carried out on January 16, 17, and August 13, 14, 2018. In each operation, the average concentrations of the fractionated tritiated water were approximately 1.5 GBq/m^3 and 1.0 GBq/m^3, respectively. The former is mainly the recovery water during the deuterium plasma experiment, the latter is diluted by the recovery water from the vacuum vessel ventilation gas which contains water vapor (H_2O) with low tritium concentration. The total amount of wastewater and tritium in the wastewater were approximately 0.85 m^3 and 1.07 GBq, respectively.

3.3. The estimation of tritium inventory and balance in EDS

The tritium inventory in EDS calculated by the summation of the tritium inventory in the MS and the PM systems and the tritium balance in EDS are shown in Figs. 4 and 5. Since some of the tritiated water recovered by EDS was fractionated into the wastewater containers, the tritium inventory in EDS decreased in January and August 2018. The tritium inventory in EDS is estimated to be approximately 1.18 GBq at the end of LHD vacuum vessel maintenance activity.

The estimation of tritium amount in the wastewater tanks is summarized in Table 2. Some of the wastewater, which contained tritium, remained in the wastewater tanks for the monitoring of water levels after fractionation operations. The volume of water in the wastewater tanks after each fractionation operation was approximately 1.1~1.2 m^3. Assuming that the tritium concentration in these wastewater tanks was the same, the tritium inventory in the wastewater after the second fractionation operation was approximately 1.1 GBq. Therefore, the main tritium inventory location in the EDS was in the wastewater tanks.

The polymer membrane used in the PM system absorbs water, however, the amount of water in the polymer material is estimated to be extremely low [14]. Thus, there would be

virtually no tritium inventory in the polymer membrane. On the other hand, the molecular sieve used in the MS system would be a part of the tritium inventory due to the presence of residual water in the molecular sieve after the regeneration operation [15-18]. Based on the regeneration operating conditions of molecular sieves [7], the amount of residual water in molecular sieves is estimated to be several kilograms. Its tritium inventory would be roughly estimated to be less than 0.1 GBq from the tritium balance in EDS at the end of the maintenance activity. When the EDS is dismantled, the tritiated water in the wastewater tanks would be easily collected at the large storage tank and would be fractionated into the wastewater containers. Thus, it would be essential to decontaminate the molecular sieve to be lower the tritium inventory in the MS system. For the decontamination of molecular sieves, the isotope swapping method using water vapor (H_2O) has been carried out [19] and was effective for decontaminating tritium in absorbent materials.

4. Summary

The exhaust detritiation system has been operated since December 2016 and then the tritium recovery operation started on March 7, 2017, for the LHD deuterium plasma experiment. The tritium produced by the D-D reaction was released from the LHD vacuum vessel walls. Released tritium released was recovered as tritiated water by EDS. The total amount of recovered tritium was estimated to be 2.4 GBq. The recovery tritiated water was dispensed into the wastewater container on January 16, 17, and August 13, 14, 2018. The amount of tritiated wastewater delivered to the JRIA was approximately 1.07 GBq. Considering the decay of tritium due to its half-life of 4500 days, the tritium inventory in the EDS is estimated to be 1.18 GBq at the end of the LHD vacuum vessel maintenance activity. Since the wastewater, which includes tritium, normally remained in the tanks for monitoring wastewater levels, the residual water in the tanks was the main tritium inventory. The molecular sieve used in the MS system would include tritiated water as the inventory due to the residual water after the regeneration operation. Its tritium inventory could be roughly estimated to be less than 0.1 GBq. Decontamination of molecular sieves would be preferred when dismantling the EDS.

Acknowledgments

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Table 1. Average tritium concentration and the amount of wastewater delivered to the JRIA.

Number of deliveries	1st	2nd
Working day	January 17 and 18, 2018	August 14 and 15, 2018
Average concentration [GBq/m^3]	1.5	1.0
Amount of wastewater [m^3]	0.45	0.40
The total amount of tritium [GBq]	~0.67	~0.40

Table 2. The estimation of tritium amount in the wastewater tanks in EDS.

Number of deliveries	Temporary receiver tank in MS system [m^3]	Temporary receiver tanks in the PM system [m^3]	Temporary large storage tank [m^3]	Wastewater measuring tank [m^3]	Average tritium concentration in wastewater containers [GBq/m^3]	Estimated tritium inventory in these tanks* [GBq]
1st	0.020	0.061	1.1	0.023	1.5	1.8
2nd	0.023	0.070	1.0	0.023	1.0	1.1

*The tritium concentration assumed to be the same in these tanks

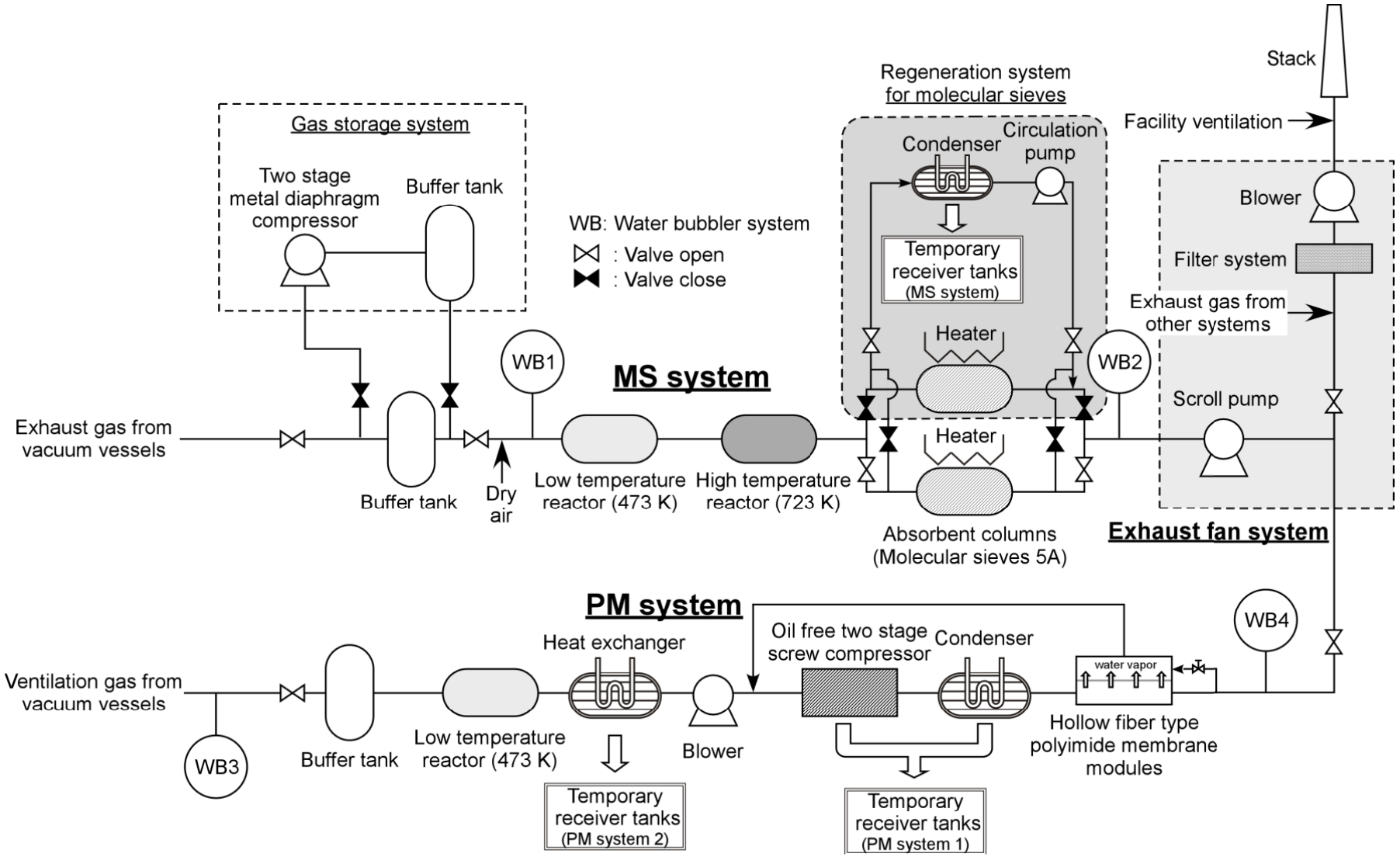

Fig. 1 Schematic diagram and process flow of the EDS and the exhaust fan system

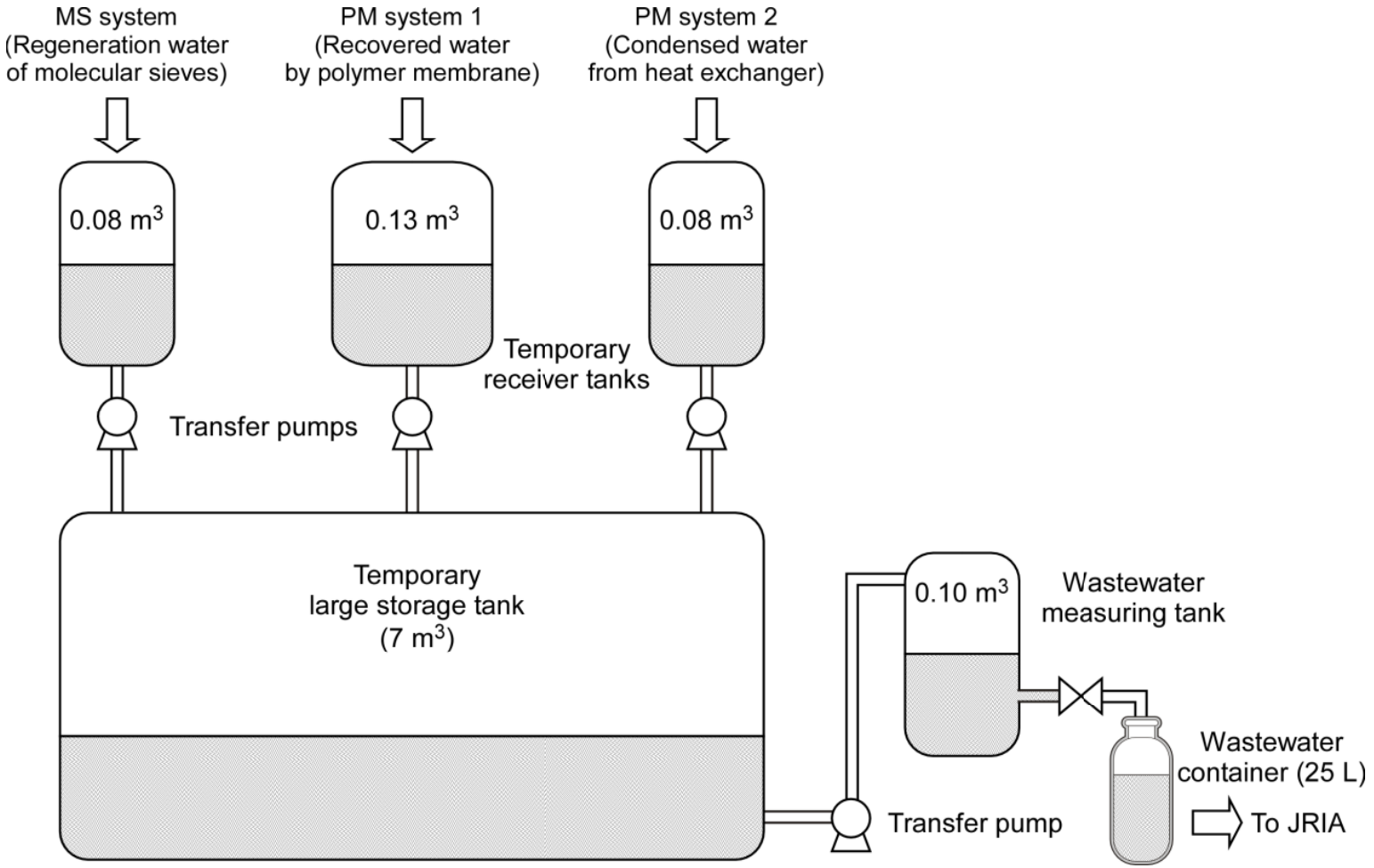

Fig. 2 The wastewater delivery and storage system in EDS

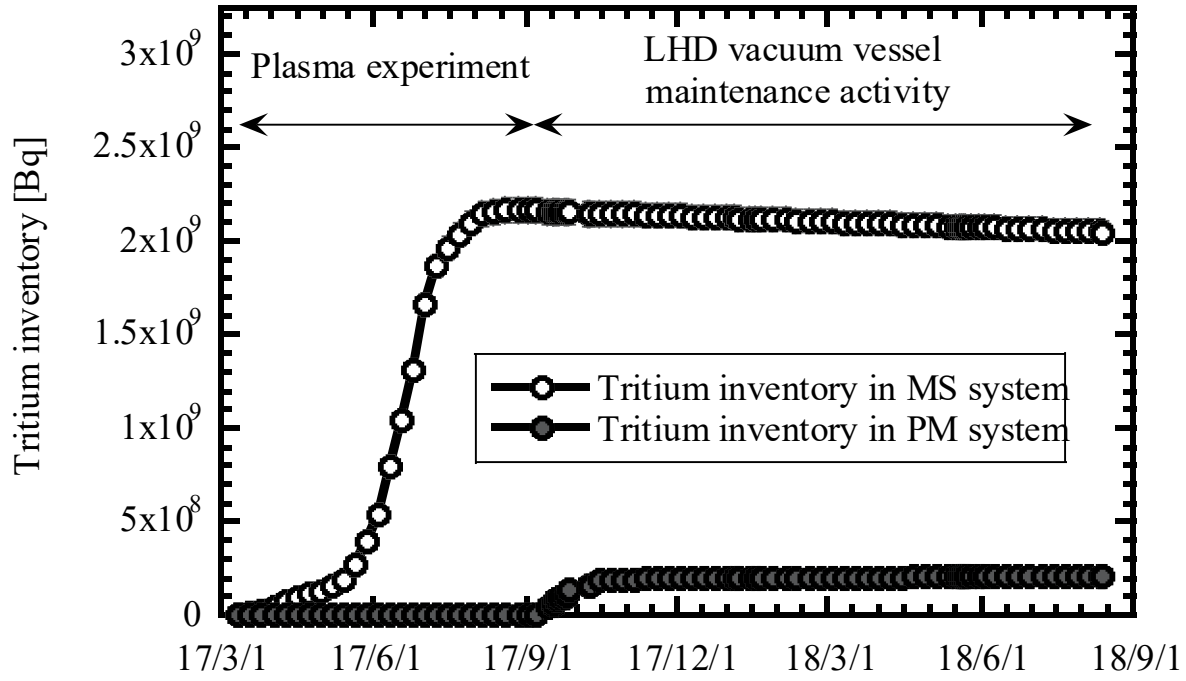

Fig. 3 The variation of tritium inventory in the MS and the PM systems.

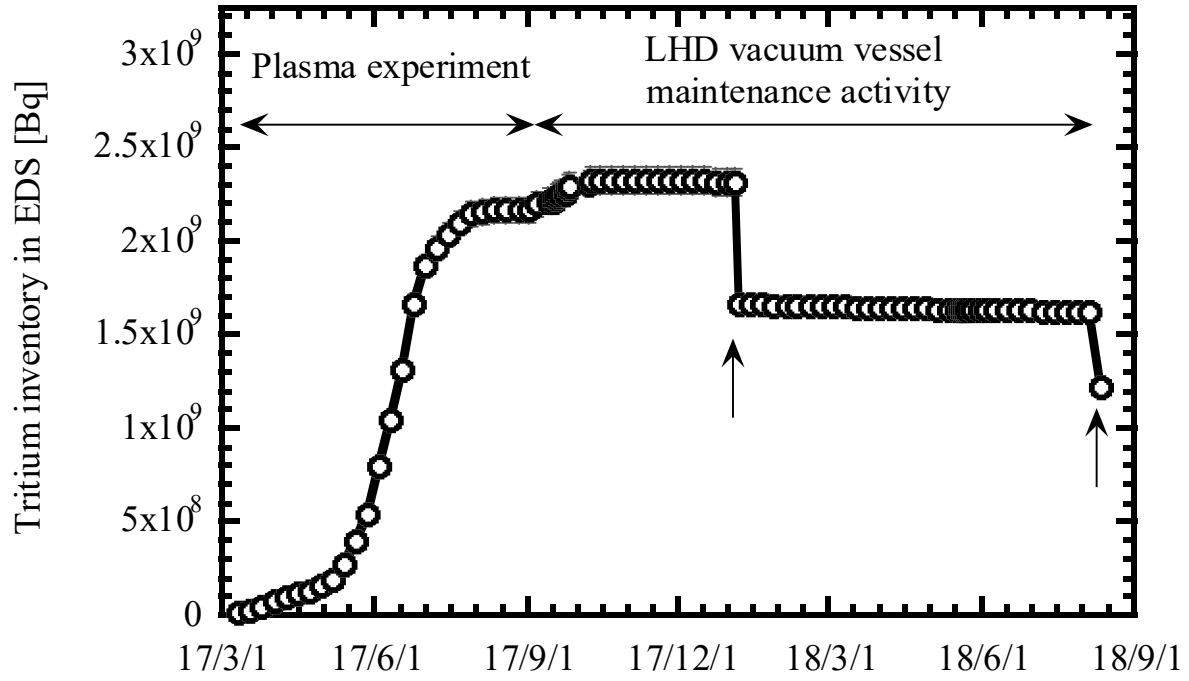

Fig. 4 The variation of tritium inventory in EDS during the plasma experiment and the LHD vacuum vessel maintenance activity. The up-arrows indicate the day of the wastewater fractionation operation.

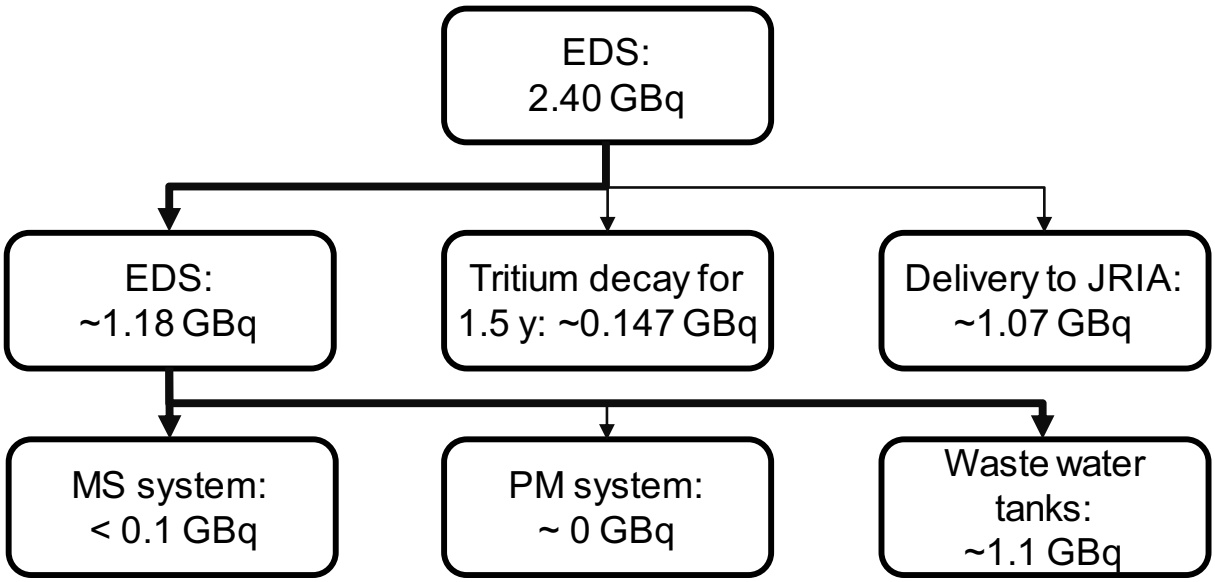

Fig. 5 The tritium balance and inventory in EDS at the end of the LHD maintenance activity in the first deuterium plasma experiment campaign.