Tritium Balance in Large Helical Device during and after the First Deuterium Plasma Experiment Campaign*)

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The Large Helical Device (LHD) started the deuterium plasma experiment on March 7, 2017. Approximately 6.4 GBq of tritium was produced in the first deuterium plasma experiment campaign and were utilized as tracers for the investigation of release behavior and the balance of tritium in the LHD vacuum vessel. To determine the tritium balance in LHD, the tritium release from the vacuum vessel was continually observed during the plasma experiment period and the vacuum vessel maintenance activities. The tritium exhaust rate was approximately 32.8% at the end of the plasma experiment. After the plasma experiment, the vacuum vessel was ventilated by room air for the maintenance activity and the tritium release from the in-vessel components was observed. The tritium release rate gradually decreased and became constant after four-month in spite of water vapor concentration. It is suggested that the tritium release mechanism from the vacuum vessel is a diffusion-limited process from the bulk. The tritium release amount during the maintenance activity for one year was approximately 5.0%. Considering the decrease of tritium decay for 1.5 years, tritium inventory in LHD was estimated to be approximately 3.66 GBq (57.2%) at the end of maintenance activity.

Keywords: Large Helical Device, deuterium plasma experiment, vacuum vessel ventilation, tritium inventory, tritium balance, tritium release behavior

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1. Introduction

Tritium (T) is one of the hydrogen isotopes and a radioactive material. It emits beta rays with the maximum energy of 18.6 keV and decays into helium-3 with the half-life of 4500 days [1]. Tritium in nature is continually produced by the nuclear reaction between the Earth's atmosphere and the cosmic rays which are composed of high-energy particles of nuclear matter in the upper atmosphere. The global equilibrium content of tritium in nature is estimated to be approximately 1.0 - 1.3 EBq [2, 3]. In the future nuclear fusion reactor, hydrogen isotopes of deuterium (D) and tritium will be utilized as fuel. The tritium inventory in the fusion reactor system will substantially exceed more than a few kg. Thus, the safe handling and radiation management of tritium in the fusion facilities are among the key issues for the public's acceptance and for the realization of a nuclear fusion reactor [4]. In the deuterium plasma experiment using a large fusion test device, a small amount of tritium is produced by the d(d, p)t reaction. The produced tritium has a high energy of 1.01 MeV and is implanted into the plasma-facing material in the vacuum vessel. Thus, although part of the produced tritium is exhausted from the vacuum vessel, the rest of the tritium would be retained in the vacuum vessel. These sources of tritium can be utilized as a tracer in a fusion test device. One of the applications is to quantitatively determine the tritium balance in the vacuum vessel. Other applications are to observe the tritium release from the fusion test device during the plasma experiment period and the maintenance activity period. As a result, the inventory and behavior of tritium in the fusion test device would be revealed. The research using tracer tritium produced by D-D reaction has been conducted by large tokamaks such as JT-60U [5–10] in the deuterium plasma phase, JET [11, 12], and TFTR [13–15] during the deuterium plasma phase prior to the D-T experiment.

At National Institute for Fusion Science (NIFS), the first deuterium plasma experiment using Large Helical Device (LHD) started on 7 March 2017 [16, 17]. After starting the deuterium plasma experiment, tritium was observed in the exhaust gas. Exhausted tritium from the LHD vacuum vessel must be recovered by the tritium removal system, such as an exhaust detritiation system (EDS) from the viewpoints of public acceptance [18, 19]. Therefore, all exhaust gas lines from LHD, Neutral Beam Injection (NBI) systems, and others had to be connected to the EDS for tritium recovery. Also, since the tritium was chronically released from the LHD vacuum vessel during the maintenance activity after the deuterium plasma experiment,

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the ventilation air passing through the LHD vacuum vessel must be treated by the EDS. Therefore the tritium recovery operation is performed throughout the year by EDS. In this study, we discuss the tritium release behavior and tritium balance in the LHD vacuum vessel during the maintenance activity after the plasma experiment.

2. Vacuum Exhaust and Ventilation System for LHD and Tritium Monitoring System

2.1 System of vacuum exhaust in LHD and ventilation during maintenance activity

A schematic of the system of LHD vacuum exhaust and ventilation during the maintenance activity is shown in Fig. 1. Because the plasma operation is performed under the vacuum condition of less than 1 Pa, LHD is equipped with various vacuum pump systems for the LHD vacuum vessel, NBI system, and plasma diagnostics, etc. The operation of the vacuum pump system was continually performed throughout the plasma experiment campaign. The hydrogen isotope gases of H_2 and D_2 were utilized as the operation gas and then hydrogen isotope gas containing tritium was exhausted from the vacuum vessel. Also, the wall conditioning operation by wall baking at 368 K and hydrogen or helium glow discharge were conducted as required. Under these operations, tritium was released from the plasma-facing components. The plasma exhaust gases containing tritium were introduced into the MS system in the EDS. The amount of tritium produced by the D-D reaction was estimated from the number of neutrons measured by the neutron flux monitor [20, 21].

After the plasma operation, the LHD vacuum vessel was opened and exposed to atmospheric pressure. First, the dependence of water vapor on the tritium release behavior from the vacuum vessel was investigated. The detailed procedure is described in Sec. 2.3. Subsequently, the workers entered the vacuum vessel for the inspection and the replacement of some of the damaged parts. Also, the test pieces for research on plasma material interaction were exchanged with new test pieces. To maintain the working environment in the vacuum vessel and to prevent oxygen deficiency among the workers, the LHD vacuum vessel was ventilated by the wet room air at the flow rate of $300 \sim 650\,\mathrm{Nm^3/h}$. Since the volume of the LHD vacuum vessel is estimated to be approximately $255\,\mathrm{m^3}$, the number of air changes per hour in the vacuum vessel becomes $1 \sim 2.5$ changes per hour. The ventilation air, which contained a small amount of tritium, was introduced into the PM system in the EDS. Thus, the inlet of EDS becomes suitable for both the observation of the behavior and the balance of tritium in LHD.

2.2 Tritium monitoring devices

The tritium monitoring devices were installed in the EDS. Regarding the MS system, the original water bubbler system [22] and the ionization chamber (Y221G0300, Ohkura Electric Co., Ltd.) were prepared for the plasma

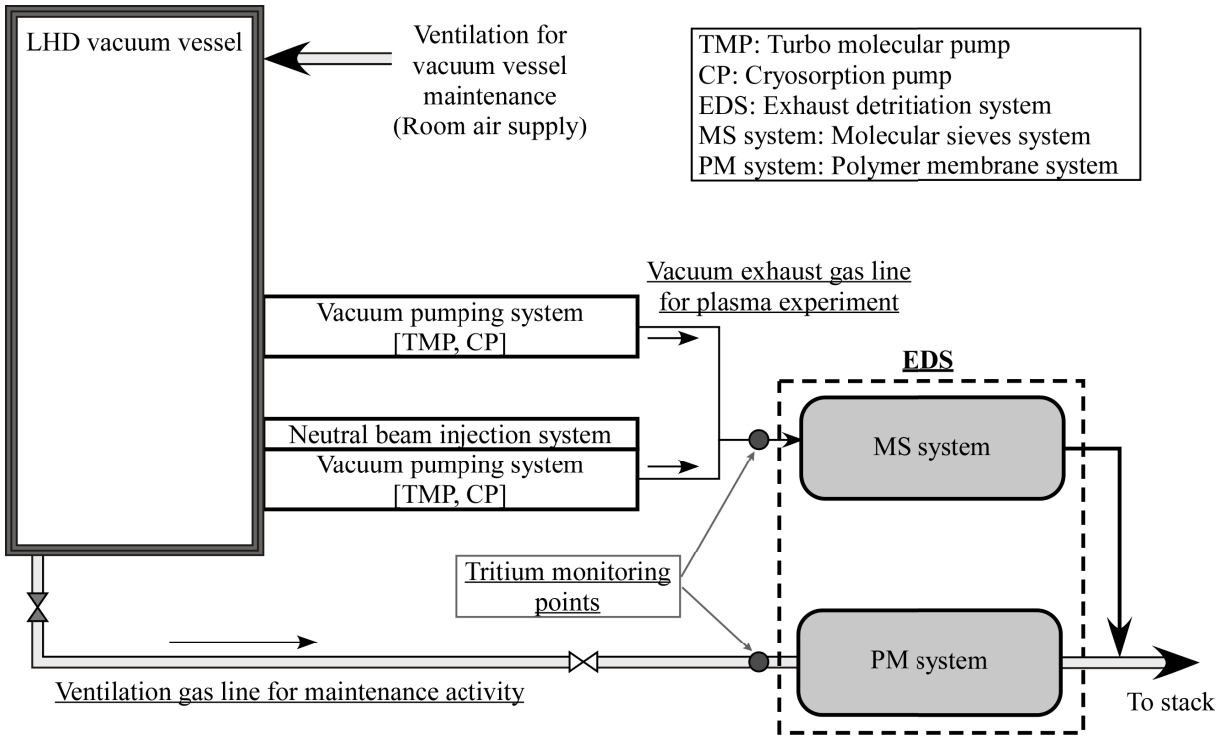

Fig. 1 A schematic of the system of the LHD vacuum exhaust and the air ventilation during the maintenance activity.

experiment. The water bubbler system is for the measurement of average tritium concentration and the discrimination of tritium chemical forms. The ionization chamber is for the real-time tritium measurement. The sampling period of the water bubbler system was one week and the sampling gas volume was less than 2 m^3. The detection limits of the water bubbler system and the ionization chamber were less than 10^{-6} Bq/cm^3 and approximately 4×10^{-2} Bq/cm^3, respectively. As for the PM system, the water bubbler system (MARC7000, SDEC France) and the proportional counter (LB-110, Berthold Technologies) were prepared for the maintenance activity in the LHD vacuum vessel. The hygrometer (Vaisala, DTM-132, measurement range: $-20°C \sim 50°C$) was installed at the inlet of the water bubbler system. The sampling period of the water bubbler system was one or two weeks and the sampling gas volume became approximately $6 \sim 9$ m^3. The detection limits of the water bubbler system and the proportional counter were approximately 10^{-6} Bq/cm^3 and 6.2×10^{-4} Bq/cm^3 at 30 minutes counting, respectively.

To determine the tritium activity in the water sample collected by the water bubbler system, the water sample (10 cm^3) was mixed with a liquid scintillator (10 cm^3, Ultima-Gold LLT, Perkin Elmer Co., Ltd.) in 20 cm^3 polyethylene vials. The background sample was prepared using deionized water in a 20 cm^3 vial. The deionized water for the background sample was the same as the ultrapure water (Direct-Q UV, Merck Millipore) used in the bubbler column. After leaving these samples for several hours in a liquid scintillation counter (Tri-Carb 4910TR, Perkin Elmer Co. Ltd.), the tritium activity was measured for a total counting time of 50 min per sample. The detection limit of the water sample was approximately 0.01 Bq/cm^3.

2.3 Tritium release behavior after plasma experiment

To confirm the mechanism of tritium release from the vacuum vessel exposed to the atmosphere, the dependence of water vapor concentration on the tritium release from the vacuum vessel was evaluated after one month from the end of the plasma experiment with reference to the results of JT-60U [9]. As the results of JT-60U, tritium in the carbon-based vacuum vessel could be easily removed by the water vapor exposure. Figure 2 shows the schematic diagram of the configuration for the experiment on tritium release from the LHD vacuum vessel. To control the dew point in the vacuum vessel, dry air and wet room air were mixed in a designated ratio. The experimental procedure was as described below, 1) The LHD vacuum vessel was evacuated at a few Pa. 2) The dry and wet room air was introduced into the LHD vacuum vessel with the designated mixing rate until reaching atmospheric pressure. 3) The LHD vacuum vessel was closed from a few hours to a few days, and the pressure inside was kept at room temperature. 4) After the expiration of a predetermined time interval, the LHD vacuum vessel was evacuated by a dry pump until a few Pa. During the time when the vacuum vessel closed off, the tritium would be released from the plasma-facing material in the vacuum vessel and be in equilibrium with the water vapor (H_2O) in the atmosphere. The experiment conditions are summarized in Table 1. The dew point and the tritium concentration in the vacuum vessel were measured continuously with a hygrometer (Vaisala, DTM-

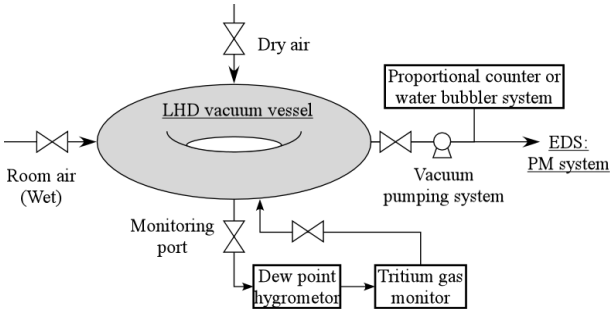

Fig. 2 The schematic diagram of the configuration for the experiment on tritium release from the LHD vacuum vessel.

Table 1 The conditions of the wet air exposure experiment in the vacuum vessel opening.

Run No.	Mixing ratio		Period	Closed time [h]	Dew point [°C]	H_2O concentration [ppm]
	Room wet air	Dry air				
Run1	0	1	2017/9/7	3.6	-59.7	11
Run2	0.03	0.97	2017/9/13-14	19.5	-43.8	82
Run3	0.1	0.9	2017/9/14-15	16.6	-26.1	560
Run4	0.8	0.2	2017/9/15-19	91.8	6.0	9685
Run5	0.2	0.8	2017/9/19-20	17.7	-14.2	1739
Run6	0.5	0.5	2017/9/20-21	16.8	-7.3	3242
Run7	0.8	0.2	2017/9/21-22	17.7	-0.4	5780
Run8	0.8	0.2	2017/9/22-25	65.5	0.9	6465

252, measurement range: $-80°C \sim 10°C$) and an ionization chamber (Hitachi Co. Ltd., DMG-233B, the detection limit of tritium: $\sim 1.1\times10^{-2}$ Bq/cm^3).

3. Tritium Behavior and Tritium Balance in LHD

As mentioned above, the first deuterium plasma experiment in LHD started on March 7, 2017. The plasma operation using deuterium gas (D_2) was carried out until June 30th, 2017. Then the plasma operation gas was changed to hydrogen (H_2) for the tritium removal in the vacuum vessel and carried out until August 3, 2017. The total amount of produced tritium was estimated to be approximately 6.4 GBq from the data of the neutron flux monitor. After the plasma experiment, the LHD vacuum vessel was opened from September 2017 and the maintenance activity of LHD was continued until August 2018. In these periods, the tritium released from the vacuum vessel was continually monitored.

3.1 Tritium release from the vacuum vessel during the plasma experiment period [23, 24]

The tritium was observed in the exhaust gas from the beginning of the deuterium plasma experiment. The tritium exhaust rate, which is defined as the ratio of exhausted tritium amount and produced tritium in the vacuum vessel, was 4.7% in the first week. The amount of exhausted tritium was gradually increased and maximized at the end of the deuterium plasma phase. The tritium exhaust rate was approximately 25.9% until the end of the deuterium plasma phase. In other words, the produced tritium was implanted in the plasma-facing components and then the tritium inventory was maximized. In the hydrogen plasma phase, the tritium was not produced by D-D reaction and the amount of exhausted tritium increased slightly. This suggested that the tritium implanted in the plasma-facing components was barely released due to the plasma discharge operation. On the other hand, tritium in the vacuum vessel was released by wall conditioning operations of hydrogen (H_2, D_2) glow discharge and wall baking at 368 K. Finally, the tritium exhausted rate was approximately 32.8% at the end of the plasma experiment.

3.2 Tritium release from the vacuum vessel during the maintenance activity

After the plasma experiment campaign ended, the vacuum vessel was opened for maintenance activity from September 2017. Before starting the maintenance activity, tritium release behavior from the vacuum vessel was evaluated as a function of water vapor concentration as described in Sec. 2.3 because it is well-known that the tritium in the vacuum vessel is released by the isotope exchange reaction with water vapor. For example, Kobayashi *et al.* have reported that the tritium contamination in the Caisson, which is 12 m^3 of gas-tight box, was removed by the isotope exchange between water vapor in the atmosphere and the adsorbed water on the wall in a short time when the water vapor was added into the Caisson [25].

Figure 3 shows the dependence of the water vapor concentration on the tritium release amount at room temperature. The tritium release amount seems to increase as the water vapor concentration increases at the initial phase from Run1 to Run4. However, when the water vapor concentration became low following the operation of Run4, the tritium release amount shifted to a much lower amount. This indicated that the tritium release amount from the vacuum vessel does not depend on the water vapor concentration during the diffusion-limited phase after the isotope exchange dominant phase at the initial vacuum vessel opening. Figure 4 shows the tritium release rate as a function of the order of the Run number. The tritium release rate was calculated by dividing the tritium release amount by closed time. The tritium release rate was decreased with the number of the Run, i.e., elapsed time after the vacuum

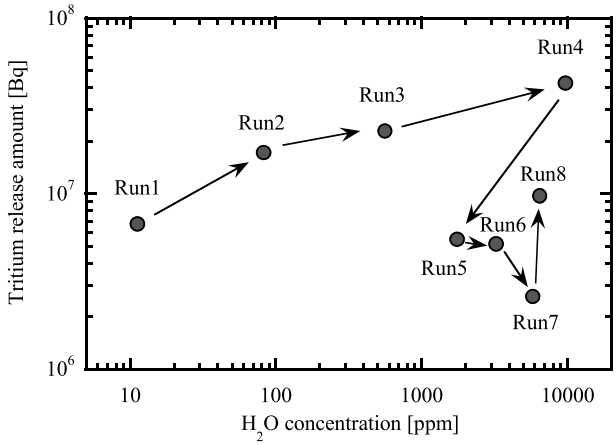

Fig. 3 The dependence of the water vapor concentration on the tritium release amount from the LHD vacuum vessel at room temperature.

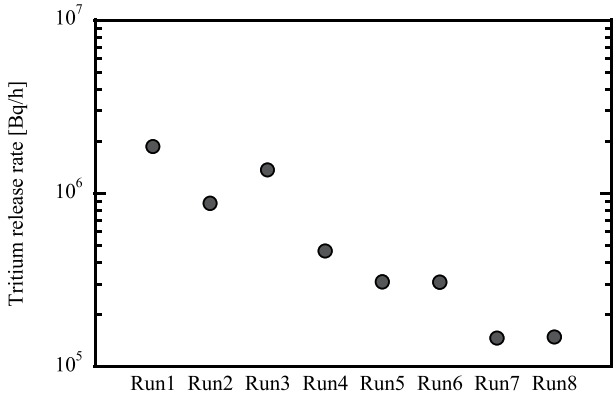

Fig. 4 The tritium release rate as a function of the order of Run number.

vessel opening. Stencel *et al.* have reported the tritium release behavior in TFTR after the vacuum vessel opening [13]. In their report, the produced tritium amount from late 1985 through July 1987 was estimated to be approximately 9.8 GBq. After the plasma experiment, the TFTR vacuum vessel was ventilated by air. The levels of tritium concentration in the TFTR vacuum vessel were decreased by air ventilation with time and were below the radiation work permit requirements of 46×10^{-3} Bq/cm^3 in PPPL after 6 days from the vacuum vessel opening. The tritium release behavior in the LHD vacuum vessel was similar to the results of air ventilation in the TFTR vacuum vessel.

Following the evaluation of tritium release dependence on the water vapor concentration, the vacuum vessel was purged by wet air for the ventilation. The variations of the tritium release rate and water vapor concentration in the ventilation air during the maintenance activity are shown in Fig. 5. The tritium release rate was high at the beginning of maintenance activity because the water vapor concentration in the ventilation air was high. The tritium on the surface of plasma-facing components could be released by hydrogen isotope exchange reaction between tritium and water vapor. However, the tritium release rate was gradually decreased according to the decrease with water vapor concentration and became almost constant after the fourth month following opening the vacuum vessel. From March 2018, although the water vapor concentration increased again, the tritium release rate was kept at constant. It indicated that the mechanism of tritium release from the vacuum vessel would be a diffusion-limited process. The diffusion-limited process on the tritium release from the vacuum vessel has been also observed during the wall conditioning operation [23]. In the deuterium plasma experiment, high energy triton less than 1 MeV is produced in the plasma and implanted into the vacuum vessel. The depth of implanted tritium in the stainless steel is estimated to be approximately a few μm. Thus, the rate-limiting factor in the tritium release behavior would be the diffusion-limited process from the bulk rather than the isotope exchange reaction on the surface.

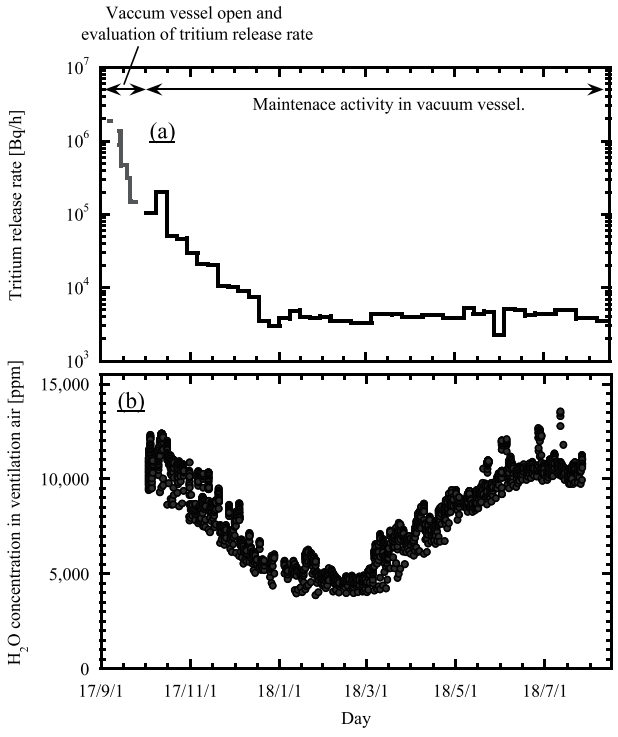

Fig. 5 The variations of the tritium release rate and water vapor concentration in the ventilation air during the LHD vacuum vessel maintenance activity: (a) tritium release rate, (b) water vapor concentration.

Figure 6 shows the variation of the exhausted tritium amount and the tritium exhausted rate during the first deuterium plasma experiment campaign and the LHD vacuum vessel maintenance activity. The tritium amount was determined by the water bubbler system. As mentioned in Sec. 3.1, the tritium release amount was maximized at the end of the deuterium plasma operation, although the tritium release amount was temporarily increased after the plasma experiment by the evaluation of tritium release. Then the tritium release amount was almost constant at the order of $10^5 \sim 10^6$ Bq until the end of the maintenance activity. The tritium exhaust rate reached 37.8% before the next plasma experiment. Figure 7 shows the variation of tritium inventory in the vacuum vessel from March 2017 to October 2018. The tritium inventory in the vacuum vessel was estimated by the following two equations:

$$A_n = A_{n-1} \times \left(\frac{1}{2}\right)^{\frac{t}{T}} + P_n - E_n, \tag{1}$$

$$A_n = A_{n-1} \times \left(\frac{1}{2}\right)^{\frac{t}{T}} - E_n, \tag{2}$$

where Eq. (1) represents the variation of the tritium inventory during the plasma experiment period, Eq. (2) repre-

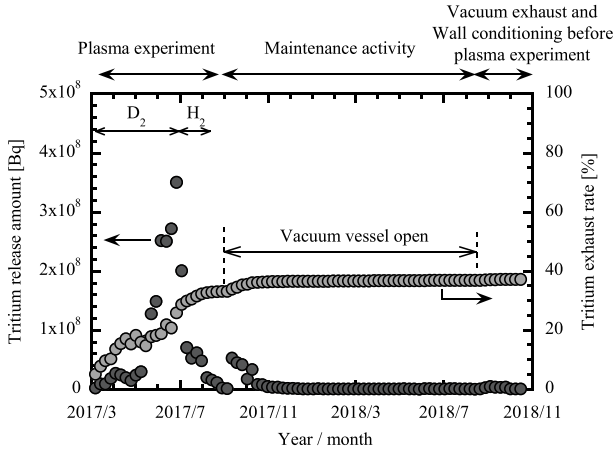

Fig. 6 The variation of the exhausted tritium amount and the exhausted tritium rate during the first deuterium plasma experiment campaign and the maintenance activity.

sents the tritium inventory during the maintenance activity. A_n is the tritium inventory at the n-th week and A_o is zero, T is the half-life of tritium, 4500 days, t is the interval of monitoring, P_n is the amount of produced tritium in the n-th week, and E_n is the amount of exhausted tritium in the n-th week. The first term on the right side in the equations represents the tritium decay during the interval of monitoring. The tritium inventory maximized at the end of the deuterium plasma experiment and was estimated to be approximately 5.0 GBq. Then, the tritium inventory in the vacuum vessel gradually decreased and became approximately 3.66 GBq before the next plasma experiment campaign. According to the calculation results of tritium decay, the amount of tritium decay for 1.5 years was estimated to be approximately 0.32 GBq.

3.3 Tritium inventory in NBI cryosorption pump

After the plasma experiment, the NBI vacuum vessel was opened. Some of the pieces of activated carbons which had pealed off the NBI cryosorption panels in the vacuum vessel were collected. The tritium activity in the activated carbons was measured by the immersion method. The sample pieces were immersed in the deionized water for a few days. The immersion water was mixed with the scintillator and then the mixed sample was measured by a liquid scintillation counter. As a result, the average tritium inventory in a piece was estimated to be 0.04 Bq. The density of activated carbons on the cryosorption panel is estimated to be approximately 30 particles/cm^2 and the area of cryosorption panel in 5 NBIs is calculated to be approximately 240 m^2. Thus, the tritium inventory in the NBI cryosorption pump was estimated to be approximately 3 MBq. This corresponds to less than 0.1% of the produced tritium amount.

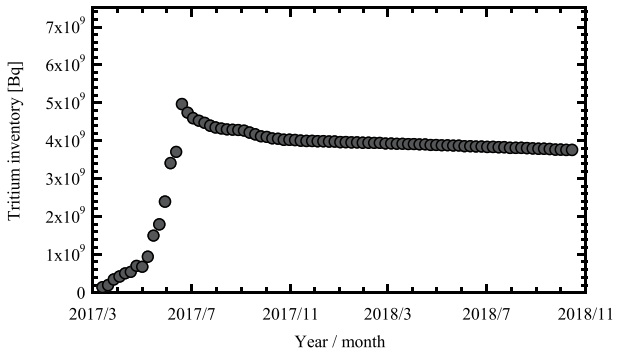

Fig. 7 The variation of tritium inventory in the LHD vacuum vessel.

3.4 Tritium in the plasma-facing materials

The tritium analysis in the plasma-facing materials has just started. The initial results of tritium distribution on the surface by using the Imaging Plate (IP) technique have been reported [26]. A relatively large amount of tritium was observed on the surfaces of the baffle part tile. On the other hand, a relatively small amount of tritium was observed at the strike point of divertor legs. As for the depth profile of tritium in the baffle part tile, larger IP intensity than the surface was observed inside the baffle part tile, and that suggests the peak of retained tritium was at several μm depth. The detailed analysis of the tritium inventory in the plasma surface materials will be conducted in the near future.

3.5 Tritium balance in LHD before next plasma experiment campaign

The diagram of tritium balance in LHD is summarized in Fig. 8. The produced tritium amount was estimated to be 6.4 GBq in the first deuterium plasma experiment campaign. A part of the produced tritium was exhausted from the LHD vacuum vessel. As mentioned in Sec. 3.2, the total tritium exhaust rate was approximately 37.8% at the

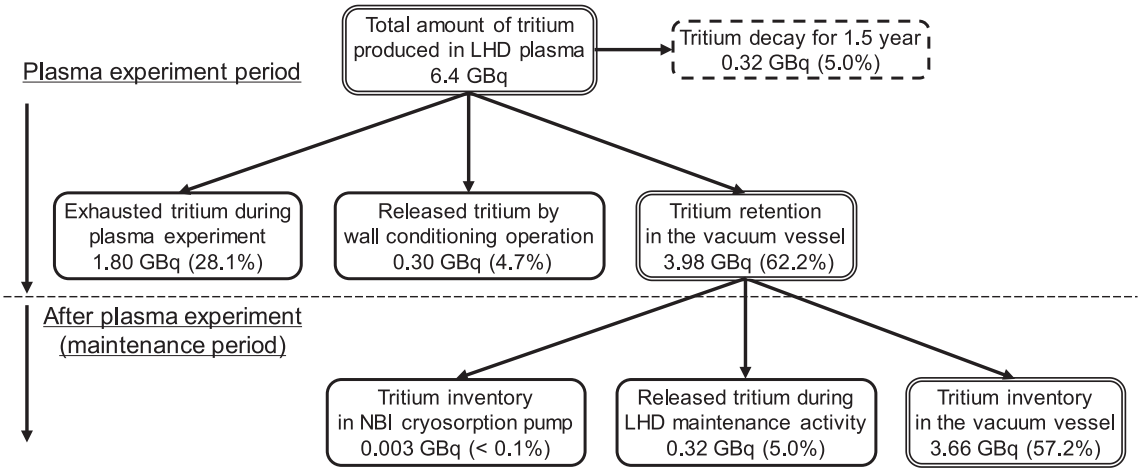

Fig. 8 The diagram of tritium balance in LHD throughout the first deuterium plasma experiment campaign and the maintenance activity.

end of the maintenance activity. The total amount of exhausted tritium consisted of the directly exhausted tritium (1.80 GBq, 28.1%), tritium release by wall conditioning operation (0.30 GBq, 4.7%) and tritium release during the maintenance activity for one year (0.32 GBq, 5.0%). The amount of tritium decay for 1.5 years from March 2017 to October 2018 was estimated to be approximately 5.0% (0.32 GBq). The tritium inventory in NBI cryosorption pumps was measured by an immersion method and estimated to be less than 0.1% (< 3 MBq). It is so small that it may be ignored in the tritium balance in LHD. Finally, the tritium inventory in LHD was estimated to be approximately 3.66 GBq (57.2%). The determination of the tritium inventory in LHD was left as a subject for future discussion. The quantitative tritium analysis of the plasma-facing components is currently underway to determine the tritium inventory in LHD.

4. Conclusion

The deuterium plasma experiment using LHD was started on March 7, 2017. A small amount of tritium was produced by the deuterium plasma and it was utilized as a tracer for the investigation of behavior and balance of tritium in the LHD vacuum vessel. To determine the tritium balance in LHD, the tritium release from the vacuum vessel was continually observed in the first plasma experiment campaign and during the vacuum vessel maintenance activity. The amount of produced tritium was approximately 6.4 GBq in the deuterium plasma experiment and the part of tritium was exhausted from the vacuum vessel. The tritium exhaust rate was approximately 32.8% at the end of the plasma experiment. After the plasma experiment, the vacuum vessel was opened and ventilated by room air for the vacuum vessel maintenance activity and then the tritium was released from the in-vessel components. The tritium release rate gradually decreased and became constant after four months in spite of water vapor concentration. It is suggested that the tritium release is a diffusion-limited process from the bulk. The tritium release amount during the maintenance activity for one year was approximately 5.0%. Considering the decrease by tritium decay for 1.5 years, tritium inventory in LHD was estimated to be approximately 3.66 GBq (57.2%) at the end of the maintenance activity. To confirm the tritium inventory in LHD, the tritium analysis on the plasma-facing materials will be conducted in the near future.

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