Control of dehumidification using polymer permeable membrane and its application to tritium removal system design

メタデータ	言語: eng 出版者: 公開日: 2021-06-23 キーワード (Ja): キーワード (En): 作成者: Tanaka, Masahiro メールアドレス: 所属:
URL	http://hdl.handle.net/10655/00012507

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Initial operation results of exhaust detritiation system using a polymer membrane

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An exhaust detritiation system using commercially available hollow fiber-type polyimide membrane modules, a PM system, was installed and applied for tritium recovery in the vacuum vessel purge gas of a large fusion test device. The PM system is operated annually and there were no serious malfunctions after starting the operation for approximately 2.5 y. The continuous recovery of tritiated water without the heating operation and the switching of valves as well as the regeneration operation of molecular sieves, was demonstrated. The tritium in the process gas was successfully recovered by the PM system even though the average tritium concentration at the inlet of the PM system was less than the order of 10^{-3} Bq/cm^3. The detritiation factor and tritium recovery ratio were greater than 10^3 and 0.97, respectively. The tritium concentration at the outlet of the PM system was maintained in the order of 10^{-6} Bq/cm^3 irrespective of the tritium concentration at the inlet of the PM system. Thus, the operating data indicates that the PM system has a potentially higher detritiation factor than DF=10^3.

Keywords: fusion test device, tritium removal, polymer membrane, exhaust gas

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1. Introduction

Tritium is a hydrogen isotope and radioactive material. Therefore, it has to be handled safely and confined to the facility. The tritium removal system, such as the air detritiation system in the room and exhaust detritiation system, play a crucial role in tritium confinement and recovery. The conventional tritium removal technique involves catalytic oxidation and water absorption using absorbents such as molecular sieves [1, 2]. The molecular sieve-type tritium removal system has been used in various tritium handling facilities.

As part of the nuclear fusion research program at the National Institute for Fusion Science (NIFS) in Japan, deuterium plasma experiments using the Large Helical Device (LHD) have been conducted to investigate high-temperature plasma physics and the hydrogen isotope effect since 2017 [3, 4]. In the deuterium plasma experiment using a large fusion test device, tritium is produced in the vacuum vessel by the deuterium–deuterium reaction. Then, the produced tritium is exhausted with both deuterium gas and hydrogen gas via the vacuum pumping system, and a part of the tritium is retained in the plasma-facing components. After the plasma experimental campaign, the vacuum vessel of the fusion test device was opened for maintenance activity and then the workers entered the vacuum vessel. To decrease the internal exposure of workers by tritium and prevent a shortage of oxygen, the vacuum vessel is ventilated by room air. The ventilated air contains tritium because the retained tritium is released from the surface of the plasma-facing components. Although the tritium concentration in ventilated air is low, tritium must be removed by the exhaust detritiation system (EDS) from the viewpoints of radiation management and public acceptance. The EDS [5] consists of two systems: the conventional molecular sieves (MS) type tritium removal system, which is operated for the tritium removal of the vacuum exhaust gas during the plasma experiment. The other is the polymer membrane (PM) type, which is used for the tritium removal during the vacuum vessel maintenance activity and of the exhaust gas of rough pumping from LHD and Neutral Beam Injections (NBIs). A tritium recovery system using a polymer membrane has been investigated and developed for a few decades [6–15]. The driving force for gas permeation is the differential pressure between the inside and outside of the polymer membrane. Thus, when the compressed wet air is fed into the inside of the polyimide membrane, the water vapor contained in the tritium is selectively permeated via the polyimide membrane [15]. Tritium recovery by the polymer membrane has the following advantages: a more compact system than the molecular sieves type system, reduced energy consumption, continuous tritium recovery operation without switching valves and heating operation, and ease of maintenance. In NIFS, the tritium recovery system using a polymer membrane has been commissioned and put to practical use since December 2016.

In this study, we describe the characteristics of continuous operation under actual conditions and the results of tritium recovery using a polymer membrane.

2. Configuration of the PM system and tritium monitoring system

2.1. PM system in the EDS

The specifications and the process flow diagram of EDS have been detailed in a previous paper [5]. In this section, we briefly describe the process flow and specifications of the PM system. A schematic of the PM system is presented in Fig. 1. This system was designed and constructed by JGC Corporation. The PM system is connected to the vacuum vessels of the LHD, 5 NBIs, and other devices. It consists primarily of an oxidation reactor for hydrogen gas combustion, a blower, a screw type compressor, a cold condenser, 20 hollow fiber-type polymer membrane modules (UM-C10, UBE Industry Ltd.), wastewater tanks, and instrumentation for moisture indication (TE-660TR, TEKHNE Corporation), vortex flow meter (digitalYEWFLO Series, Yokogawa Corporation) and tritium monitoring system. Although the main process gas is wet air containing tritiated water vapor, trace hydrogen gas might be contained in the process gas. Therefore, an oxidation reactor packed with a platinum catalyst is installed, which oxidizes trace hydrogen gas in the process gas. The temperature of the reactor was maintained at 200 °C. After passing through the reactor, the process gas is compressed to more than 0.6 MPa(G), and the moisture in the process gas is condensed by both the aftercooler in the compressor and cold condenser. Subsequently, the rest of the moisture is separated by the polymer membrane modules. The separated moisture is returned to the inlet of the compressor and condensed by the cold condenser again. As a result, the moisture is continually recovered as tritiated water. The condensed tritiated water is received in the temporary receiver tank, after which it is transferred to the temporary storage tank when the liquid level in the temporary receiver tank attains a high level.

The specifications of the PM system are summarized in Table. 1. The maximum process flow was designed to be 300 Nm^3/h in order to ventilate the large-volume vacuum vessel of the LHD. This system is operated continuously for approximately 8,400 h annually. The remainder annual usage time is scheduled for the system maintenance that is required for proper operation. The tritium recovery ratio is required to be more than 95% according to the agreement with local governments [16]. The tritium recovery ratio, R, is defined as:

$$\begin{aligned} \mathrm{R} &= \left(1 - \frac{\mathrm{C_{out}} \times \mathrm{F_{out}}}{\mathrm{C_{in}} \times \mathrm{F_{in}}}\right) \\ &= \left(1 - \frac{\mathrm{C_{out}}}{\mathrm{C_{in}}}\right) \\ &= \left(1 - \frac{1}{DF}\right), \qquad (1) \end{aligned}$$

where C_{in} and C_{out} are the tritium concentration at the inlet and outlet of the PM system, respectively. Further, F_{in} and F_{out} are the flow rates at the inlet and outlet of the PM system, respectively. Although some of the process gas in EDS is refluxed as shown in Fig. 1, the overall flow path is once-through. Therefore, the flow rate at the inlet, F_{in}, and the outlet, F_{out}, is the same. *DF* is the detritiation factor, defined as the ratio of C_{in} and C_{out}.

2.2. Tritium monitoring

The tritium concentration in the process gas was monitored by a proportional counter (LB-110, Berthold Technologies GmbH & Co.) and a water bubbler system for tritium sampling (MARC 7000, SDEC France). These instruments were installed at the inlet of the PM system. The counting time of the proportional counter is 30 min, and the detection limit is approximately 0.62–0.70 mBq/cm^3. The water bubbler system is operated for a period between one day and two weeks according to the operation constraints of the PM system. The sampling gas volume was 0.5–1.0 m^3. The sample water containing tritium was mixed with the scintillation solution (UltimaGold LLT, Perkin Elmer) and then measured by a liquid scintillation counter (Tri-Carb 4910TR, Perkin Elmer). The total counting time was 50 min. The detection limit of the tritium concentration in the process gas is of the order of 10^{-6} Bq/cm^3.

The tritium concentration at the outlet of the PM system was monitored by an original water bubbler system [17]. This system is operated with the same sampling period as the tritium sampler system at the inlet of the PM system. The tritium concentration in the sample water was determined by the liquid scintillation counter. The detection limit is of the order of 10^{-6} Bq/cm^3.

3. Operation results of the PM system

The PM system has been operated since December 2016 before the LHD deuterium plasma experiment was started and cannot be shut down except during maintenance. Annual maintenance of the system was conducted to ensure proper operation, for which the operation was shut down for two weeks. A recovery performance test using hydrogen gas was carried out after the annual maintenance. So far, a hydrogen recovery ratio of 95% or more, which is the required specification, has been achieved, without deterioration of system performance.

The total operating time was more than 23,000 h until September 2019. Since the start of operation in December 2016, there have been no serious malfunctions that have caused the PM system to shut down due to component failures for a period of time that is longer than a day.

3.1. Continuous water recovery operation

The polymer membrane in the PM system can separate tritiated water vapor. The tritiated water is recovered continuously. Figure 2 illustrates the variation of the process parameters during the continuous water recovery operation one night. The process gas was wet air, which ventilated the LHD vacuum vessel. The water recovery operation was conducted from 17:00 to 8:30. The water in the temporary receiver tank was transferred to the temporary storage tank at 20:00 because the liquid level in the temporary receiver tank attained a high level. The process flow and water vapor concentration in the process gas were 300 Nm^3/h and 1.1%, respectively. The recovery ratio of water vapor was greater than 99%. The total amount of water recovered during the operation was calculated to be approximately 40 L from the experimental data in Figs. 2(a) and (b). The rate of increase of recovery water was estimated to

be constant at 2.6 L/h. Thus, the liquid level in the temporary receiver tank increased at a constant rate, as depicted in Fig. 2(c). The total amount of recovery water was estimated to be approximately 42 L from the liquid level data in the temporary receiver tank. The amount of water corresponds to the calculated value. Therefore, these experimental results indicated that continuous water recovery by the PM system was successfully achieved.

3.2. Recovery operation with an impurity gas

During the maintenance activity period, the residual gas in the gas distribution system was released into the purge gas line. The released operational gases were hydrogen isotope gas (H_2, D_2), nitrogen gas, and noble gases such as helium gas. The hydrogen isotope gas was oxidized by the catalyst and converted into water vapor. Therefore, inert gases as impurity gases were introduced into the polymer membrane modules with wet air as the process gas. The polyimide membrane module can separate the various gases according to the process conditions. The gas permeability of PI at 60^{o}C °C is summarized in Table 2. Among these inert gases, helium gas has a high gas permeability, and the selectivity over nitrogen gas is more than 160. Thus, helium gas is separated from the process gas by the polyimide membrane module. Figure 3 shows the variation in pressure at the outlet of the compressor and the flow rates in the membrane module system. The viscosity of helium gas is almost the same as that of air, and thus the effect of helium gas on the measurement by the vortex flow meter is expected to be practically negligible. The permeated flow rate was calculated by subtracting the residual flow rate from the feed flow rate. Helium gas was introduced into the process gas of wet air from 10:30 to 12:00. The pressure was temporally decreased during the helium gas supply because the helium gas had a low density. On the other hand, the permeated flow rate increased despite the pressure drop. The high permeability of helium would increase the permeated flow. Since the permeated helium gas was not recovered by the condenser, helium gas may be gradually released into the residual gas. It becomes a normal permeated flow rate after the helium gas supply operation.

In this operation, the residual gas flow rate was decreased by the helium gas supply. When the volume of helium gas is larger than the condition specified by this operation, the residual gas flow rate may fall below the lower limit of the PM system, thereby stopping the system automatically. Therefore, the process gas containing helium as the impurity gas is unsuitable for this type of tritium recovery system using a polyimide membrane module. It is necessary to redesign the process flow diagram to successfully achieve tritium removal from the helium gas.

3.3. Operation results of tritium recovery

The PM system is a backup system for the MS system during the plasma experiment. Thus, tritium removal by the PM system was carried out at the beginning of the maintenance activity after the plasma experiment. Figure 4 shows an example of the variation of the tritium concentration at the inlet of the PM system during the LHD and NBI vacuum vessel purge

operation. The tritium concentration was measured using a proportional counter. These vacuum vessels were closed in an atmosphere with moisture at night. Then, the purge operation was conducted from 10:00 to 15:00 for NBI and from 15:00 to 19:00 for LHD. The water vapor concentration in the purge gas was approximately 0.50–0.85%. The recovery ratio of water vapor was more than 97%. The tritium concentration in the NBI vacuum vessel purge gas was less than 10^{-3} Bq/cm^3. This indicated that tritium was not retained in the NBI vacuum vessel. The tritium concentration in the LHD purge gas operation temporally increased because the produced tritium was implanted in the vacuum vessel and released from the plasma-facing components during the night. The maximum tritium concentration was 7.6×10^{-2} Bq/cm^3. Then, it was gradually decreased until the end of the purge operation because tritium in the LHD vacuum vessel was purged and replaced by room air. On the other hand, the tritium concentration at the outlet of the PM system was too low to be measured by the proportional counter. Thus, a water bubbler system was installed, and the average tritium concentration was less than 10^{-3} Bq/cm^3. The results of the tritium removal operation are summarized in Table 3. The tritium removal operations were conducted from 13–25th March 2019. The average tritium concentrations at the inlet of the PM system were in the order of 10^{-3} Bq/cm^3 and 10^{-5} Bq/cm^3. The tritium concentrations at the outlet of the PM system were kept on the order of 10^{-6} Bq/cm^3 during these operations. The tritium recovery ratio in these operations was more than 0.97. The PM system could recover tritium in the process gas and satisfy the required tritium recovery ratio, as detailed in Table 1, even though the tritium concentration at the inlet of the PM system was extremely low.

The maximum detritiation factor (DF) achieved was 1475 under these operating conditions. It was dependent on the tritium concentration at the inlet of the PM system because the concentration at the outlet of the PM system was kept low regardless of the concentration at the inlet. Therefore, the tritium removal system using a polymer membrane may be capable of decontaminating tritium by more than DF=10^3 in case of higher tritium concentration. During the tritium removal operation, the dew point at the outlet of the PM system was also kept low (below -45 ^{o}C) regardless of the water vapor concentration in the inlet. On the other hand, although the tritium concentration at the outlet of the PM system during the tritium removal operations was kept less than 10^{-5} Bq/cm^3, it gradually increased. One of the reasons might be the backward flow contained tritium from the downstream of the PM system. Because the inlet valve of the PM system is closed when the tritium removal operation is not conducted and there is no gas flow in the PM system. During the non-tritium removal operating period, some of the gas that contained tritium was released directly into the downstream pipelines of the PM system, and a part of the gas stayed in the pipeline. At this time, the tritium in the downstream may have diffused and was collected by the water bubbler system at the outlet of the PM system because the dew point at the outlet of the PM system was slightly increased. Consequently, the tritium concentration at the outlet may have increased.

4. Conclusion

The exhaust detritiation system using a polymer membrane, PM system, has been operated since December 2016 for the LHD deuterium plasma experiment. Commercially available hollow fiber-type polyimide membrane modules were applied to the polymer membrane in the PM system. The PM system mainly treats the vacuum vessel purge gas containing tritium. It operates all year-round except for the stop by the maintenance work. There were no serious malfunctions after starting the operation.

The tritium in the process gas was successfully recovered by the PM system even though the average tritium concentration at the inlet of the PM system was less than the order of 10^{-3} Bq/cm^3. This system demonstrated the continuous recovery of tritiated water without any heating operation and switching the valve, like the regeneration of molecular sieves. The detritiation factor and tritium recovery ratio were greater than 10^3 and 0.97, respectively. The tritium concentration at the outlet of the PM system was kept constant on the order of 10^{-6} Bq/cm^3 during the tritium recovery operation. Thus, these operating data indicate that the PM system has a potentially higher detritiation factor.

Acknowledgments

This work was supported by the NIFS budget NIFS18ULAA708 and ULAA023, and partially by JSPS KAKENHI Grant Number 17K06998. We wish to thank Ms. C. Iwata of NIFS for her technical support during the course of this work.

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Table 1. Specifications of the PM system in EDS.

System	Polymer membrane type
Process gas	Purge gas from LHD and NBI vacuum vessel and maintenance room
Max. process flow rate [Nm^3/h]	300
Gas composition in the process flow	Wet air
Detritiation factor (Tritium recovery ratio)	> 20 (> 95%)
Annual operating hours [h]	~8400 (24 h × 350 days)

Table 2. Gas permeability of polyimide at 60 ˚C

Gas	Gas permeability [18-20] (10^{-10} cm^3(STP)·cm/cm^2/s/cmHg)	Selectivity over N_2
H_2O	3100	41000
He	12	160
O_2	0.577	7.6
N_2	0.076	1
CO_2	2.7	36
CH_4	0.037	0.49

Table 3. Results of the tritium recovery operation.

Run No.	Sampling period*		Average tritium concentration		Recovery rate [-]	DF [-]	Purged vacuum vessel
	Start	End	Inlet of EDS [Bq/cm^3]	Outlet of EDS [Bq/cm^3]			
Run 1	2019/3/13	2019/3/14	9.45×10^{-5}	1.56×10^{-6}**	0.98	61	NBI
Run 2	2019/3/14	2019/3/15	5.79×10^{-5}	1.61×10^{-6}**	0.97	36	NBI
Run 3	2019/3/15	2019/3/18	2.56×10^{-3}	1.74×10^{-6}	1.00	1475	NBI/LHD
Run 4	2019/3/18	2019/3/19	2.78×10^{-3}	2.80×10^{-6}	1.00	993	NBI/LHD
Run 5	2019/3/19	2019/3/20	1.03×10^{-3}	4.34×10^{-6}	1.00	238	NBI/LHD
Run 6	2019/3/20	2019/3/22	2.89×10^{-4}	7.25×10^{-6}	0.97	40	NBI/LHD
Run 7	2019/3/22	2019/3/25	2.96×10^{-4}	8.28×10^{-6}	0.97	36	NBI/LHD

* The time of the sampling start and stop was at 9:00.

** Tritium concentration was the detection limit.

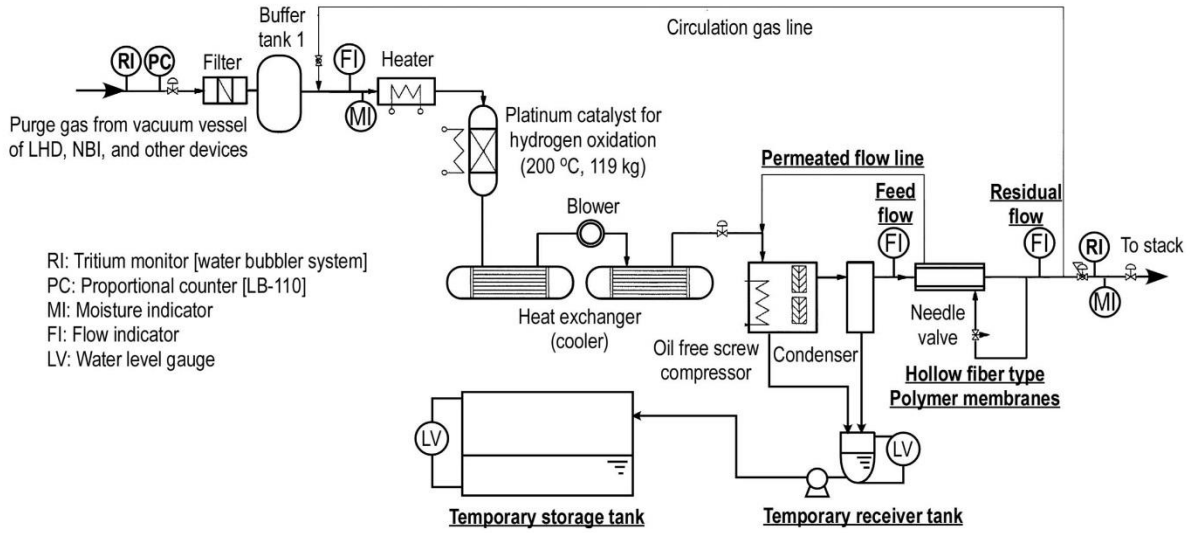

Fig. 1 Schematic diagram of the PM system.

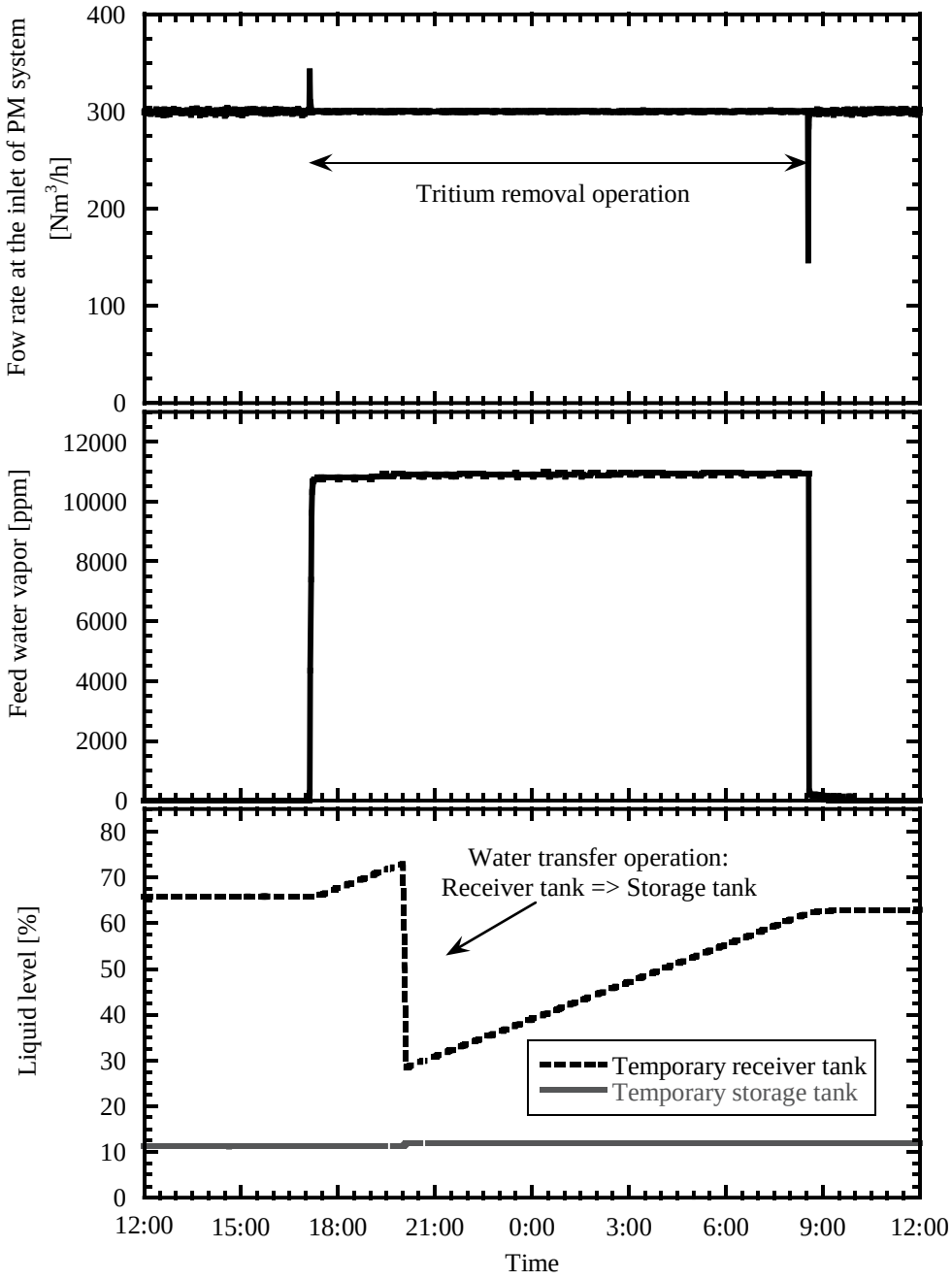

Fig. 2 The variation of the process parameters during the continuous water recovery operation for one night; (a) Flow rate at the inlet of oxidation reactor, (b) Water vapor concentration in the feed gas, (c) Liquid level in the temporary receiver tank and temporary storage tank.

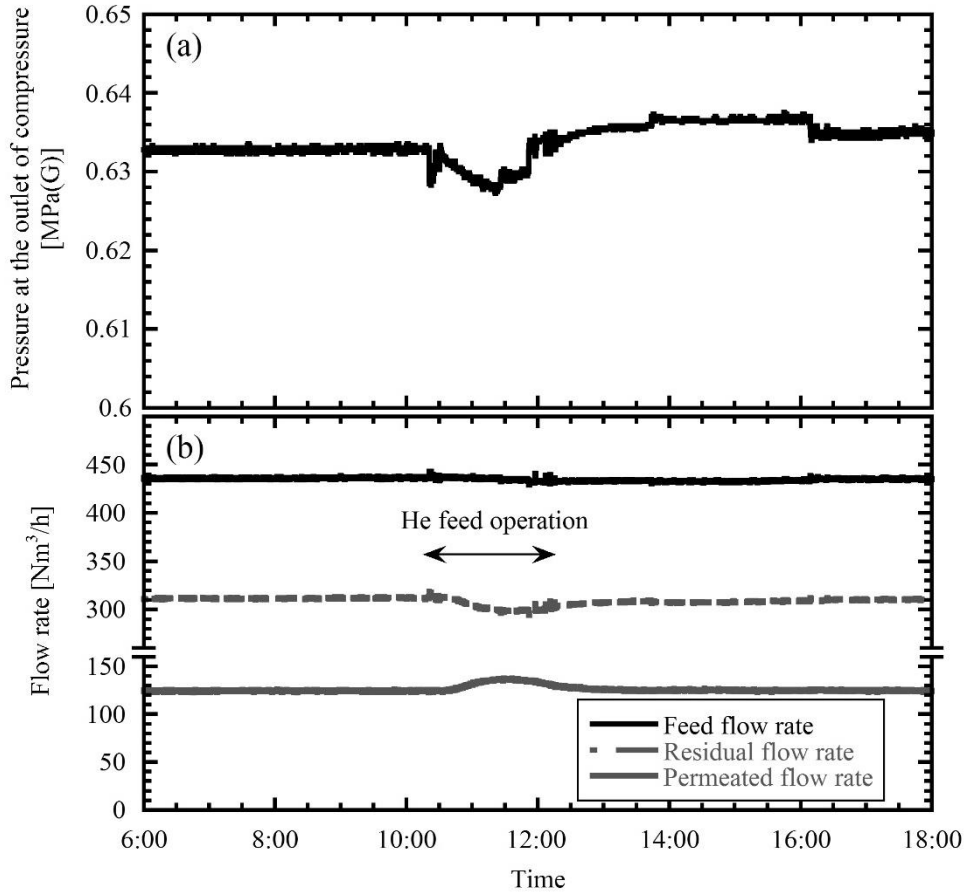

Fig. 3 Variation of (a) the pressure at the outlet of compressor and (b) the flow rates in the membrane module system during the helium gas supply.

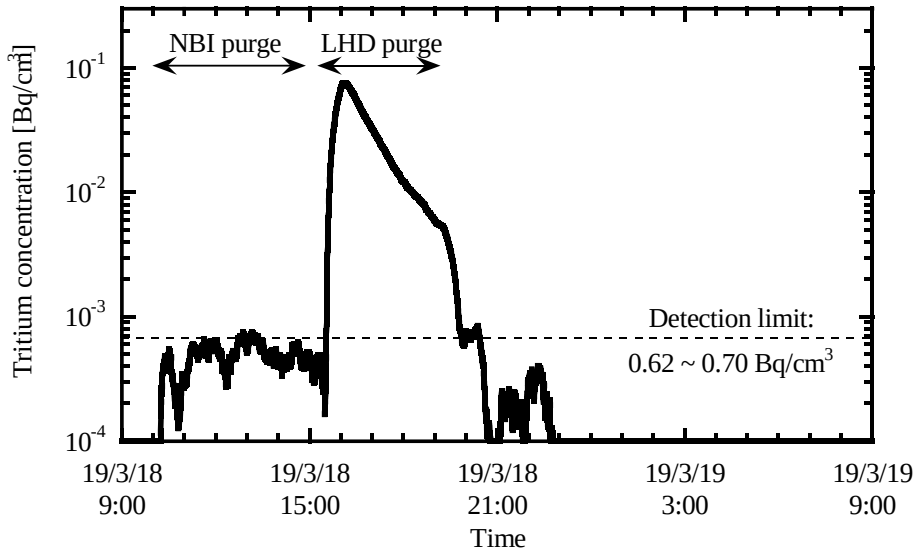

Fig. 4 An example of the variation of the tritium concentration at the inlet of the PM system during LHD and NBI vacuum vessel purge operation in a day.