Performance of exhaust detritiation system for a fusion test device in the initial phase of the operation

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The exhaust detritiation system (EDS) has been operating for the deuterium plasma experiment in large fusion test device since 2016. The EDS consists of two systems: the molecular sieves (MS) type for plasma exhaust gas, the polymer membrane (PM) type for vacuum vessel purge gas during the maintenance activity. The tritium removal performance of the EDS was evaluated for four years. As the operation results, the maximum detritiation factor (DF) of the MS system was achieved to be more than 10^3 even though the average tritium concentration in the process gas was less than 10^4 Bq/m^3. On the other hand, the DF of the PM system was less than 10^3, because the average tritium concentration during the vacuum vessel maintenance activity was at the order of less than 10^3 Bq/m^3. Throughout the initial phase of the deuterium plasma experiment, the tritium concentration at the outlet of EDS was maintained to be less than the order of 10^2 Bq/m^3. Also, a failure analysis was performed to evaluate the reliability of the EDS in the initial phase of the operation. Over the four years of operation, the overall failure rate of the MS system was on the order of 10^{-5} events/hour.

Keywords; fusion test device, tritium removal, exhaust gas, detritiation factor, failure analysis

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1. Introduction

On March 7, 2017, Large Helical Device (LHD) at the National Institute for Fusion Science (NIFS) put the deuterium plasma experiment into operation for the research of high-temperature plasma [1]. In the deuterium plasma experiment using a large fusion test device, a small amount of tritium is produced by D-D fusion reaction in the vacuum vessel and is exhausted via the vacuum pump system. Because the tritium is a radioactive material, it must be removed by the tritium removal system from the viewpoints of radiation safety and radiological control. The tritium removal system has been installed and operated in the large fusion test facilities [2-4] and the tritium handling facilities all over the world [5-8]. The conventional tritium removal technique is catalytic oxidations and water absorption using absorbent such as molecular sieves [9, 10]. In NIFS, the exhaust detritiation system (EDS) as the tritium removal system has been commissioned and operated since 2016 prior to starting the LHD deuterium plasma experiment [11]. The EDS is applied with two types of tritium recovery systems; one is 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. Other is the novel tritium recovery system using polymer membrane (PM) 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). After starting the deuterium plasma experiment, a part of the produced tritium was exhausted from the vacuum vessel during the plasma experiment [12] and was removed from the exhaust gas by the MS system. Then, a part of retained tritium in the LHD vacuum vessel was released during the vacuum vessel maintenance activity after the plasma experiment [13]. The released tritium was treated by the PM system. The preliminary results of the tritium recovery operation by EDS has been reported [14, 15]. In this report, we described the performance of tritium decontamination and the results of failure mode analysis of EDS for four years.

2. Exhaust Detritiation System and tritium monitoring system

2.1. EDS

The EDS has been constructed by JGC Corporation and commissioned in 2016. The required specifications of EDS are summarized in Table. 1 [11, 16]. The annual operation time for the MS system is approximately 4800 hours during the plasma experiment. The maximum flow rate of the MS system was designed to be 20 Nm^3/h. On the other hand, the PM system is operated throughout the year. The PM system is used as the backup system of the MS system when the plasma experiment is carried out. After the plasma experiment, the vacuum vessels are opened and ventilated by room air. Tritium is released from the surface of the LHD vacuum vessel. Thus, the ventilation gas from the vacuum vessel contains tritium and must be treated by the PM system. The maximum flow rate of the PM system was designed to be 300 Nm^3/h. The detritiation factor of both systems is required more than 20 [16].

A schematic diagram of EDS is shown in Fig.1. The MS system mainly consists of a low-temperature reactor packed with a platinum catalyst (473 K), a high-temperature reactor

packed with a palladium catalyst (723 K), two absorbent columns packed with molecular sieves 5A, and a gas storage system for the regeneration operation of NBI cryosorption pumps. The molecular sieves weight packed per one column is 416 kg. The gas storage system comprises a two-stage diaphragm compressor and a large volume buffer tank (approximately 15 m^3). For the combustion of hydrogen gas, the dry air is introduced in the inlet of the low-temperature reactor at the constant flow rate, because the vacuum exhaust gas from LHD is oxygen-free. When the flow rate at the inlet of the MS system exceeds the threshold, the main process gas line in the MS system is switched to the PM system. The PM system consists of a low-temperature catalyst (473 K) packed with a platinum catalyst, a blower, a two-stage screw type compressor, a cold condenser, 20 hollow fiber type polymer membrane modules (UM-C10, UBE Industry Ltd.), and an absorbent column packed with molecular sieves 5A. The absorbent column is used for the rough evacuation operation of the vacuum vessel after maintenance. In other systems, a wastewater delivery and storage system, and a chilled water circulation system consisting of a chiller and liquid delivery pump are installed as common utility facilities. The detailed process system and operation mode of EDS are described elsewhere in [11]. Downstream of the EDS is connected to an exhaust fan system that comprises a filter system, a blower, and a scroll pump as shown in Fig. 1. The exhaust fan system is responsible for blowing the gases treated by the EDS to the stack as well as the exhaust gases from other exhaust systems.

2.2. Exhaust gas monitoring system

To evaluate the tritium removal performance, some of the tritium monitoring systems are installed in EDS. The main tritium monitor is the original water bubbler system [17] and four water bubbler systems are installed at the inlet and outlet of the MS and the PM systems, respectively. The water bubbler system was operated from one day to two weeks according to the operation of the EDS. The sampling gas volume of the water bubbler system was controlled to be 0.5 ~ 1 m^3. The volume of sample water in one bubbler column was approximately 20 g. The sample water used for the bubbler system was deionized water produced by an ultrapure water system. After the collection operation, the sample water of 10 cm^3 is mixed with the scintillation solution of 10 cm^3 (UltimaGold LLT, Perkin Elmer) in the Teflon vial of 20 cm^3 and then measured by a liquid scintillation counter (Tri-Carb 4910TR, Perkin Elmer). The total counting time is 50 minutes. The detection limit of tritium concentration in the process gas becomes an order of 1 Bq/m^3.

As for the real tritium measurement in the process gas, an ionization chamber (Y221G0300, Ohkura Electric Co., Ltd.) for the MS system and a proportional counter (LB-110, Berthold Technologies GmbH & Co.) for the PM system are installed at the inlet of each system. The volume of the ionization chamber is 10 L and the operating pressure is 0.098 MPa (G). The sampling gas flow rate is 10 L/min. The specification for the tritium detection limit is approximately 7.0 kBq/m^3. The volume of the proportional counter is 1.3 L. The sampling gas flow rate is 0.2 L/min. The counting time of the proportional counter is 30 minutes and the detection limit becomes approximately 0.62~0.70 kBq/m^3.

3. Operation results of the EDS

The EDS has been in continuous operation since December 2016 before starting the LHD deuterium plasma experiment. So far, three plasma experimental campaigns using deuterium (D_2) and hydrogen (H_2) have been conducted, the first plasma experiment was from February 8, 2017, to August 3, 2017, the second was from October 23, 2018, to February 21, 2019, and the third was from October 3, 2019, to February 6, 2020. The amount of tritium produced in each plasma experimental campaign was 6.4 GBq, 6.0 GBq, and 2.2 GBq. After the plasma experimental campaign, the annual maintenance of EDS was carried out in August for the MS system and common utility facilities and in March for the PM system. The total suspension period for EDS due to annual maintenance is approximately three weeks. The total operating time is more than 24,000 hours for the MS system and 32,000 hours for the PM system and the utility facilities until September 2020.

3.1. The tritium removal performance of the MS system

After the start of the MS system operation, the maximum tritium concentration of 1.3 x 10^6 Bq/m^3 was observed by the ionization chamber at the inlet of the MS system. On the other hand, since the tritium concentration at the outlet of the MS system was low and could not be measured by the ionization chamber, the average tritium concentration measured by the water bubbler system was used to evaluate the tritium removal performance. The results of the tritium removal operation of the MS system is summarized in Table 2. Detritiation factor (DF) is defined as the ratio of the tritium concentration at the inlet and outlet of the MS system. In the third experiment, the LHD plasma operation was suspended for inspection from October 15, 2019, to November 11, 2019. The average tritium concentrations measured by the water bubbler system are an order of 10^4 Bq/m^3 at the inlet of the MS system and seem to depend on the amount of produced tritium in each plasma experimental campaign. The average tritium concentration at the outlet of the MS system in the first deuterium experiment was high. In the absorbent used for tritium removal operation, there are reversibly absorbed tritiated water and irreversibly retained tritium water [18]. This tritium causes the tritium memory effect of the adsorbent [19, 20]. In the MS system [11], depending on tritium recovery operating conditions, the adsorption period of one MS column is about 1 to 2 months. The regeneration period of the MS column is several days, including the cooling period. The absorbent is regenerated at 623 K in the closed-loop system which consists of a heater, a blower, a condenser at 283 K. The reversible absorbed tritiated water is released from the absorbent during the regeneration operation. The desorbed tritiated water vapor from the absorbent is almost condensed back to a liquid by the condenser and then the rest of the tritiated water vapor is recovered in the absorbent again at the end of the regeneration operation. The residual tritiated water in the absorbent after the regeneration operation is estimated to be 6~7 kg from the water adsorption isotherm data. Also, irreversibly tritium water is retained more strongly by chemical bonding in the zeolite structure. Therefore, this tritiated water in the absorbent is tritium contamination. It is known

that the outlet gas from the contaminated absorbent has a specific activity close to that of the water adsorbed during the previous absorption cycle [21]. Then, such residual tritiated water would impair the performance of the MS system. To reduce the residual tritiated water in the absorbent, the effect of isotope swamping on the tritium decontamination of the absorbent was evaluated. The decontamination procedure is as follows: 1. Introduce hydrogen gas (H_2) into the inlet of the low-temperature reactor to generate water vapor (H_2O); 2. Adsorb the water vapor onto the adsorbent; 3. Regenerate the adsorbent; 4. Repeat this procedure three times. The tritium decontamination result is shown in Fig. 2. The amount of H_2O used for the isotope swamping was almost the same as the mass of residual Q_2O (Q=H, D, T) in the absorbent. Repeating the decontamination procedure reduced the tritium concentration at the outlet of the MS system. By repeating the decontamination operation three times, the initial tritium concentration was reduced to less than one-tenth. Isotope swamping using H_2O is effective in the tritium decontamination of absorbent materials. Thus, since the second deuterium plasma experiment, the tritium concentration at the outlet of the MS system decreased compared to the first one due to the decontamination operation of the absorbent. As a result, the average decontamination factor of the MS system during the plasma experiment was maintained above 10^3, which exceeds the required specification of 20.

3.2. The tritium removal performance of the PM system

The results of the main tritium removal operating conditions of the PM system are summarized in Table 3. Since the location of the water bubbler system at the outlet of the PM system was changed in June 2018, the data measured thereafter are shown in the table. The tritium concentration at the inlet of the PM system was less than 1 kBq/m^3, which is the detection limit of the proportional counter, for most of the maintenance period, and it is difficult to evaluate the tritium removal performance. Therefore, the evaluation of tritium removal performance was carried out during the following periods; 1. Ventilation gas in the vacuum vessel after the plasma experiment, 2. Ventilation gas during the vacuum vessel maintenance activity at the early stage of the vacuum vessel opening, and 3. Rough evacuation of the vacuum vessel after maintenance. The average tritium concentration at the inlet of the PM system during the maintenance period was about two orders of magnitude lower than that in the exhaust gas during the plasma experimental period. On the other hand, the tritium concentration at the outlet of the PM system was close to the lower limit of detection. In conditions where tritium concentrations can be detected, the detritiation factor of the PM system exceeds the required specification of 20. At present, the memory effect of the membrane module has not been observed, which might be due to the low tritium concentration.

3.3. Failure analysis of EDS in the initial phase of the operation

There have been a variety of accidents and incident events that have caused the system to shut down in the initial phase of the operation. The most common event was an operational error by the operators in the early stages of operation. Operational errors were reduced by

revising the operation manual and training operators. The EDS was also shut down by an alarm signal from the integrated radiation monitoring and interlock system [22]. Among the critical events that lead to system shutdown, the failure of system components is the most critical. Therefore, the failure events and failure rate in the initial phase of operation are summarized. Table 4 shows the results of failure events in EDS since December 2016. The failure rate was calculated using the operation time of 24,000 hours for the MS system and 32,000 hours for the PM system and the utility facilities. The critical failure events that led to the MS system shutdown were those related to valve opening and closing operations. In the MS system, there is a weekly change of operation mode, at which time the valves are opened and closed. A valve opening and closing failure occurred when the operating mode of the MS system was switched. The frequency of these events was two during the initial phase of operation. Here, the failure rate λ is defined as follow;

$$\lambda = \frac{\text{Number of the failure event}}{\text{Total operation time}}.$$

Thus, the total failure rate of the critical events that led to the MS system shutdown was 8.3 x 10^{-5} events/hour in this system. These failure events could be addressed by repairing the parts and reviewing the settings. The downtime on each event was several hours, including the time to investigate the failure factors. Other failure events were not critical and allowed the operation to continue. These devices where the failure event occurred were repaired, maintained, or replaced with parts during the maintenance period of EDS. On the other hand, the valve opening and closing failures in the PM system have not occurred. In the PM system that continuously removes tritium, the operating modes do not frequently switch and thus the failure rate due to valve opening and closing would be lower than that in the MS system.

4. Summary

To remove tritium in the exhaust gas from the large fusion test device, the exhaust detritiation system (EDS), which mainly consists of the MS system for the exhaust gas during the plasma experiment and the PM system for the ventilation gas of the vacuum vessel during maintenance activity, has been operating since December 2016. The tritium removal performance of the EDS was evaluated for four years. As a result, both the MS and the PM systems performed above the required detritiation factor of 20. The detritiation factor of the MS system was reduced due to the tritium memory effect of the adsorbent after the first deuterium plasma experiment, but isotope swamping method by water (H_2O) addition decontaminated the adsorbent and improved the detritiation factor. Also, a failure analysis was performed to evaluate the reliability of the EDS in the initial phase of the operation. Over the four years of operation, the overall failure rate of the MS system was on the order of 10^{-5} events/hour.

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Credit Author Statement

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Table 1. Specifications of the EDS [11, 16].

System	Molecular sieves type (MS type)	Polymer membrane type (PM type)
Process gas	Exhaust gas from vacuum vessels of LHD, NBI, etc.	Ventilation gas from vacuum vessels of LHD, NBI, etc.
Max. process flow rate [Nm^3/h]	20	300
Gas composition in the process flow	H_2, D_2, He, N_2, Ne, water vapor, hydrocarbons, CO, CO_2, tritiated gas	Wet air
Detritiation factor (Tritium recovery rate)	> 20 (> 95%)	
Annual operating hours [h]	~4800 (24 h × 200 days)	~8400 (24 h × 350 days)

Table 2. Results of tritium removal operation by MS system

Evaluation period	Deuterium plasma experiment	Produced tritium [GBq]	Average tritium concentration		DF*		Remarks
			Inlet of EDS [Bq/m^3]	Outlet of EDS [Bq/m^3]	Maximum [-]	Average [-]	
2017/3/6 ~ 2017/8/7	1st	6.4	3.1×10^4	1.7×10^2	3.9×10^5	5.5×10^4	
2018/10/22 ~ 2019/2/25	2nd	6.0	3.5×10^4	2.2×10^1	7.0×10^3	2.3×10^3	
2019/9/30 ~ 2020/2/10	3rd	2.2	1.8×10^4	2.6×10^1	5.0×10^3	1.2×10^3	2019/10/15 ~ 11/11: Suspension of plasma experiment for inspection

*DF: Detritiation factor

Table 3. Results of tritium removal operation by PM system.

Evaluation period	Operation mode*	Average tritium concentration		Decontamination factor	
		Inlet of EDS [Bq/m^3]	Outlet of EDS [Bq/m^3]	Maximum [-]	Average [-]
2018/8/22 ~ 2018/8/31	3	1.0×10^2	1.4×10^0	7.4×10^1	7.4×10^1
2019/3/8 ~ 2019/4/1	1	7.1×10^2	6.6×10^0	4.9×10^2	1.8×10^2
2019/8/14 ~ 2019/9/9	1, 2, 3	3.4×10^2	4.8×10^0	1.0×10^2	7.4×10^1
2019/10/15 ~ 2019/11/11	1, 2, 3	1.8×10^2	2.3×10^0	1.4×10^2	8.6×10^1
2020/2/26 ~ 2020/3/9	1	9.3×10^2	4.1×10^0	1.1×10^3	6.0×10^2

* 1. Ventilation gas in the vacuum vessel after the plasma experiment,

2. Ventilation gas during the maintenance activity in the vacuum vessel at the early phase of the vacuum vessel opening,

3. Rough evacuation of the vacuum vessel after maintenance.

Table 4. Failure events in EDS since December 2016

Failure events	Failure mode	System	Frequency	Failure rate* [events/hour]	Effect and criticality**	Downtime for action***	Remarks
Valve opening/closing failure	Valve opening/closing sensor failure, valve opening failure	MS	2	8.3×10^{-5}	C	b	
Metal diaphragm compressor start-up failure	Internal interlock due to lack of operation condition	MS	1	4.2×10^{-5}	B	b	Not a major impact on operation because of the start-up phase
Thermometer anomaly	Temperature transducer failure	MS	1	4.2×10^{-5}	B	b	Less critical thermometer
Auxiliary heater failure	Heater wire breakage	MS	1	4.2×10^{-5}	B	c	Keeping the operating temperature by the main heater
Chiller shut down failure	Deviation from operating conditions	Utility facilities	3	9.4×10^{-5}	B	a	Keeping the operating temperature by the buffer water tank (0.5 m^3)

* Operation time; MS system: 24,000 hours, PM system and utility: 32,000 hours

**A: No impact, B: minor failure (operation), C: major failure (critical failure)

*** a: operation, b: less than a few hours, c: less than a few days, d: less than a few weeks, e: less than a few months

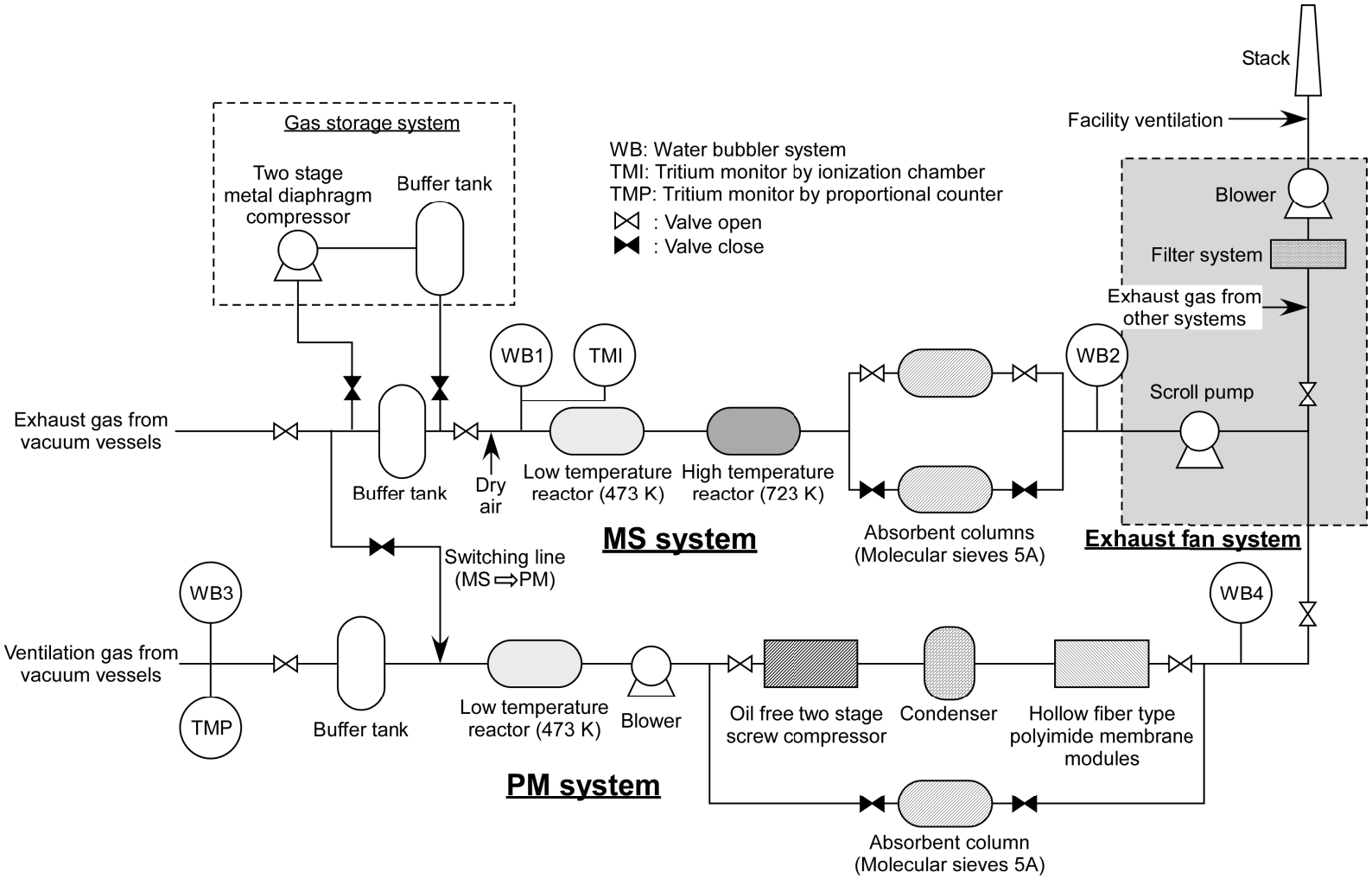

Fig. 1 Schematic diagram and process flow of the EDS and the exhaust fan system

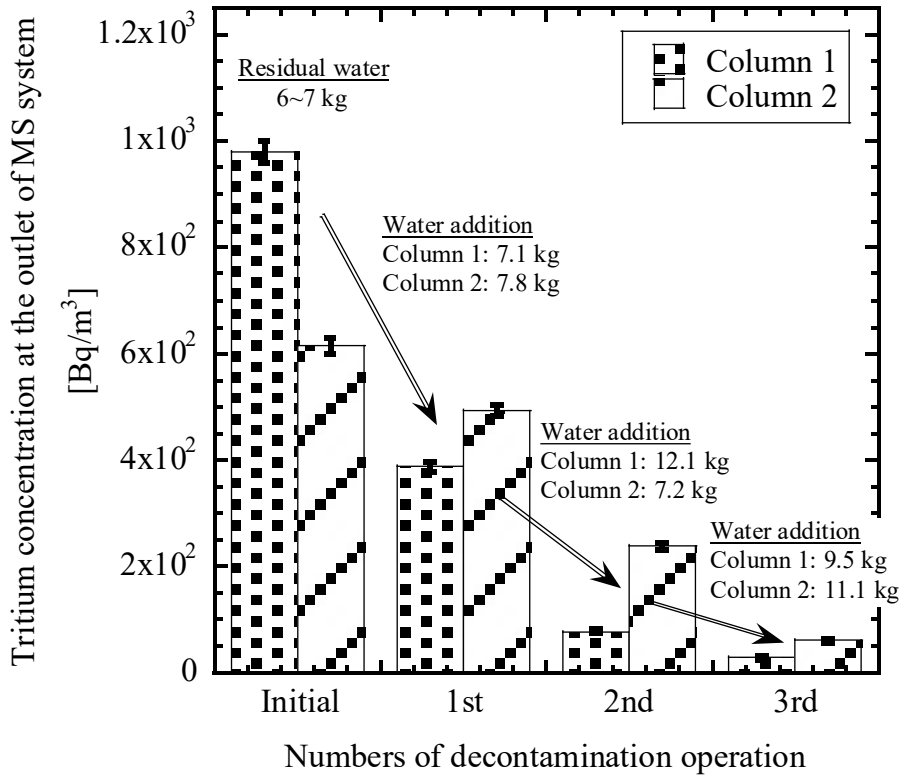

Fig. 2 The tritium decontamination of absorbents by the addition of water (H_2O) as isotope swamping.