ELSEVIER

Fusion Engineering and Design 61–62 (2002) 575–583

Fusion Engineering and Design

www.elsevier.com/locate/fusengdes

Behavior of tritium in the TSTA test cell combined with operation of the Experimental Tritium Cleanup (ETC) system

R.S. Willms [b,*], K. Kobayashi [a], Y. Iwai [a], T. Hayashi [a], S. O'hira [a], M. Nishi [a], D. Hyatt [b], R.V. Carlson [b]

[a] *Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195, Japan*
[b] *Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

Abstract

Tritium and deuterium are expected to be the fuel for the first fusion power reactors. Being radioactive, tritium is a health, safety and environment concern. Room air tritium clean systems can be used to handle tritium that has been lost to the room from primary or secondary containment. Such a system called the Experimental Tritium Cleanup (ETC) systems is installed at the Tritium Systems Test Assembly (TSTA) at Los Alamos National Laboratory. The ETC consists of (1) two compressors which draw air from the room, (2) a catalyst bed for conversion of tritium to tritiated water, and (3) molecular sieve beds for collection of the water. The exhaust from this system can be returned to the room or vented to the stack. As part of the US–Japan fusion collaboration, on two separate occasions, tritium was released into the 3000 m^3 TSTA test cell, and the ETC was used to handle these releases. Each release consisted of about one Curie of tritium. Tritium concentrations in the room were monitored at numerous locations. Also recorded were the HT and HTO concentrations at the inlet and outlet of the catalyst bed. Tritium surface concentrations in the test cell were measured before and at a series of times after the releases. Surfaces included normal test cell equipment as well as idealized test specimens. The results showed that the tritium became well-mixed in the test cell after about 45 min. When the ETC was turned on, the tritium in the TSTA test cell decreased exponentially as was expected. The test cell air tritium concentration was reduced to below one DAC (derived air concentration) in about 260 min. For the catalyst bed, at startup when the bed was at ambient temperature, there was little conversion of tritium to HTO. However, once the bed warmed to about 420 K, all of the tritium that entered the bed was converted to HTO. Immediately after the experiment, surfaces in the room initially showed moderately elevated tritium concentrations. However, with normal ventilation, these concentrations soon returned to routine levels. The data collected and reported here should be useful for planning for the operation of existing and future tritium facilities.

Keywords: Tritium; Experimental Tritium Cleanup system; Tritium System Test Assembly

1. Introduction

Tritium and deuterium are expected to be the fuel for the first fusion power reactors. Being radioactive, tritium is a health, safety and environ-

* Corresponding author. Tel.: +1-505-667-5802; fax: +1-505-667-2730
E-mail address: willms@lanl.gov (R.S. Willms).

0920-3796/02/$ - see front matter
PII: S0920-3796(02)00292-2

ment concern. Thus, considerable effort is made to ensure that tritium is always contained within process systems and secondary containment such as gloveboxes. This combination of systems will contain routinely handled tritium, and will deal with routine permeation out of the primary containment and occasional leaks of tritium out of the primary containment. An infrequent occurrence is the loss of tritium from the secondary containment into the room. It is a point of debate regarding how such releases should be handled. One point of view is that a room cleanup system should be installed to capture the tritium released to the room. The opposing view advocates direct stacking of tritium released to the room, noting that room cleanup systems require tritium to be converted to water (HTO) which is $>20\,000$ times more hazardous than elemental tritium (HT). With the decontamination factor of such systems being approximately 1000, there is plenty of room for debate.

In any event, such systems have been installed in some of today's tritium facilities. Such a system has been installed at the Tritium Systems Test Assembly (TSTA) at Los Alamos National Laboratory (LANL). This system called the Experimental Tritium Cleanup (ETC) system has been described previously, Carlson et al. [1]. A similar system is being operated at the Joint European Torus, Sabathier et al. [2].

Recently two experiments were performed to (1) study the behavior of tritium released in the TSTA test cell and (2) to test the ETC performance in responding to these releases. This paper will report on the results of these tests.

The behavior of 1 Ci of tritium released into the 3000 m^3 TSTA test cell has been reported previously, Hayashi et al. [3]. In those studies the tritium was removed from the room using the normal stack blower which generated a flow-rate of approximately 18 000 m^3/h. Elsewhere the behavior of tritium released into a 12 m^3 volume in the Caisson Assembly for Tritium Safety Study at the Japan Atomic Energy Research Institute (JAERI) has also been studied (Hayashi et al. [4], Iwai et al. [5,6], Kobayashi et al. [7]).

The present work was performed as collaborative work between LANL (US Department of Energy) and JAERI.

2. Experimental

Three tritium release experiments were performed previously, Hayashi et al. [3]. These were designated as TRE1, TRE2 and TRE3. For the present two tests the experiments were designated as TRE/ETC1 and TRE/ETC2.

2.1. ETC description

A simplified view of the ETC is given on Fig. 1. The ETC consists of (1) two compressors which draw air from the room, (2) a catalyst bed for conversion of tritium to tritiated water, and (3) molecular sieve beds for collection of the water. The actual path used for this test is shown by the bold lines on Fig. 1. As shown, only one of the ETC compressors were used for the test. The compressor used was the larger of ETC compressors, which is rated at approximately 2800 m^3/h (STP). To avoid tritium contamination of the ETC molecular sieve beds, they were bypassed for this test. The exhaust from the ETC can be returned to the room or vented to the stack. For this test the ETC was exhausted to the stack. The ETC catalyst is not heated electrically. Rather it is heated using the heat of compression from the compressor—thus the back pressure valve shown is necessary for this purpose.

For preventive maintenance purposes, the ETC was routinely operated throughout the life time of TSTA. However, the ETC had never been operated with tritium (other than the small amount of routine tritium in the test cell) prior to these tests.

2.2. Release apparatus

Essentially the same release apparatus was used for all tritium release experiments (Fig. 2). The section of tubing between two valves was loaded with nominally 1 Ci (37 GBq) of tritium (TRE3 used a larger volume so D_2 could be mixed with the tritium). This was placed between two pneu-

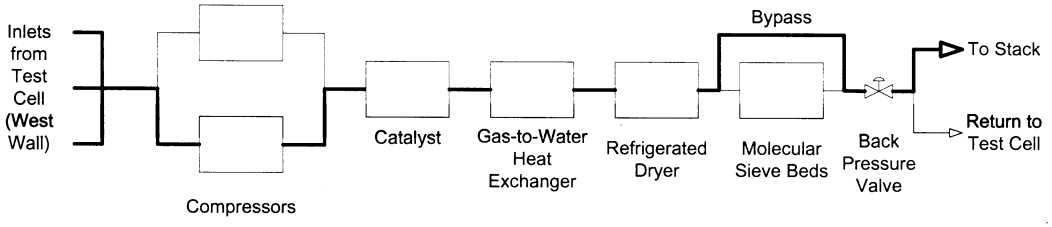

Fig. 1. Simplified view of the ETC, bold lines show the path used for this test.

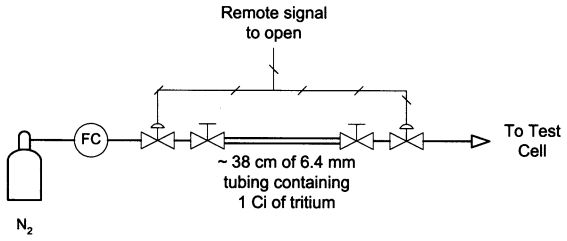

Fig. 2. Tritium release apparatus.

matically-actuated valves (closed) and the manual valves were opened. The tritium could then be released remotely by sending a signal to the pneumatic valves. Nitrogen was used to completely flush the tritium into the room at a controlled rate using a mass flow controller.

2.3. Catalyst performance

The catalyst performance was monitored by directly sampling the catalyst bed exhaust with a bubbler and an ion chamber. These two measurements determined the HTO and the total tritium leaving the bed. By difference the amount of HT leaving the bed was determined. The amount of tritium entering the bed was known by the room tritium monitor (ion chamber) readings.

2.4. Test cell monitoring

Figs. 3 and 4 are pictures of the TSTA test cell looking south and north, respectively. The pictures were taken immediately prior to TRE/ETC2. Seven ion chambers are positioned throughout the test cell for normal radiation protection. The TSTA tritium stack is monitored by an ion chamber and a bubbler. During the TRE/ETC,

Fig. 3. South-looking view of TSTA test cell.

Fig. 4. North-looking view of TSTA test cell.

two additional room monitors (ion chambers) were positioned in the test cell along with a multi-vessel bubbler with time-sequenced switching.

2.5. Surface tritium concentrations

Elevated tritium levels in the room air could lead to increased tritium surface concentrations. To quantify this, swipe samples were collected before the tritium release and for a number of days following the release. Swipes were taken of both 'ideal', specially placed materials and normal test cell surfaces. The amount of tritium collected on the swipes was determined by scintillation counting.

2.6. General procedure

The overall steps used for the TRE/ETC experiments were:

1) Prepare the test cell for the experiment including fitting it with exposure materials.
2) Prepare the tritium release apparatus.
3) Shutdown the normal room ventilation.
4) Release the tritium.
5) Monitor room concentrations using ion chambers and bubblers.
6) Start the ETC, venting the gas to the stack.
7) Continue to monitor room concentrations. Additionally monitor the ETC catalyst performance.
8) When the room tritium concentrations are reduced below 20 μCi/m^3, turn off the ETC and restore normal room ventilation.
9) Perform swipe samples over a period of days to measure surface contamination levels.

3. Results

3.1. TRE/ETC1

TRE/ETC1 was performed on 8 November 2000. The ventilation to the room was stopped at 09:58:30 h and the tritium was released at 10:00:20 h. The tritium concentrations in the room versus time after the release test are given on Fig. 5. As shown, the tritium concentrations in the room rose rapidly after the release. In about 45 min all of the tritium levels equilibrated. The differences in the tritium monitor readings are believed to be due to instrument inconsistencies rather than actual tritium concentration differences. With no ventilation the room levels decreased. This is believed to be due mostly to leaks in the test cell. Three hours after the release the ETC was started, resulting in an exponential decrease in the room concentrations. The ETC was run for about 5.75 h. During this time the room was reduced below the derived air concentration level of 20 μCi/m^3. Thereafter the normal room ventilation was restored. This returned the room to the background levels observed before the test.

3.2. TRE/ETC2

TRE/ETC2 was performed on 20 March 2001. The ventilation to the room was stopped at 08:09 h and the tritium was released at 08:30 h. All of the previous TRE and TRE/ETC tests had turned the ventilation off immediately prior to the release. The TRE/ETC2 tritium concentrations in the room versus time after the release test are given on Fig. 6. The results are very similar to TRE/ETC1. Again the ETC was started 3 h following the release. In this case the ETC was run for just over 5 h. During this time the room was reduced below the derived air concentration level of 20 μCi/m^3.

The room tritium concentrations for all five TRE and TRE/ETC experiments, showing only one room monitor reading for each experiment, are shown on Fig. 7. The TRE experiments held the tritium in the room for 5 h before reactivating the normal room ventilation. All five experiments released nominally 1 Ci of tritium. Again absolute reading differences are probably due mostly to instrument differences. The results for all experiments are quite similar other than the expected observation that the 18 000 m^3/h stack blower removed the tritium much more quickly than the 2800 m^3/h compressor.

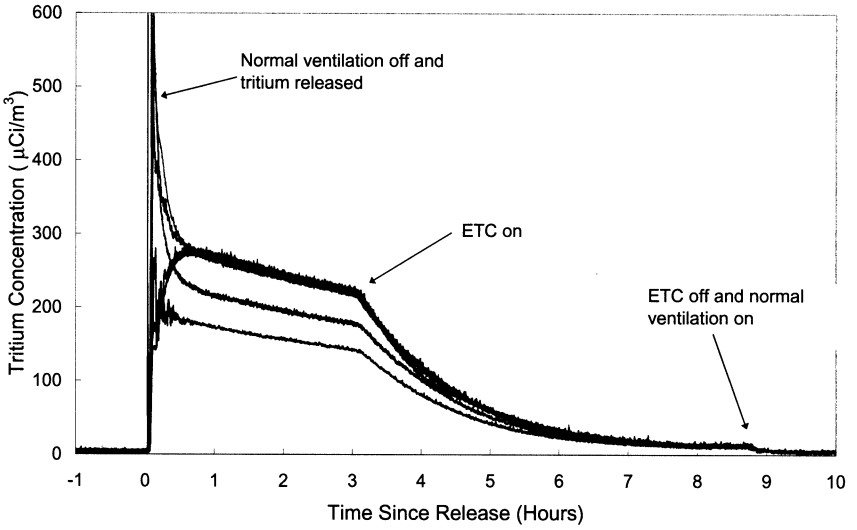

Fig. 5. Tritium concentrations (seven ion chambers) vs. time after release for TRE/ETC1.

3.3. Catalyst performance

The catalyst bed performance for both TRE/ETC experiments is summarized in Fig. 8. The tritium concentration at the exit of the catalyst bed is observed to decrease continuously over the 300 min following the start of the ETC. The catalyst bed temperature in each case starts at about 300 K and warms (by heat of compression) over a period of about 200 min to 430 K. A bubbler was used to determine the amount of HTO at the exit of the catalyst bed. Unlike ion chambers, which read instantaneously, bubblers collect samples over a period of time. Thus, the horizontal bars on Fig. 8

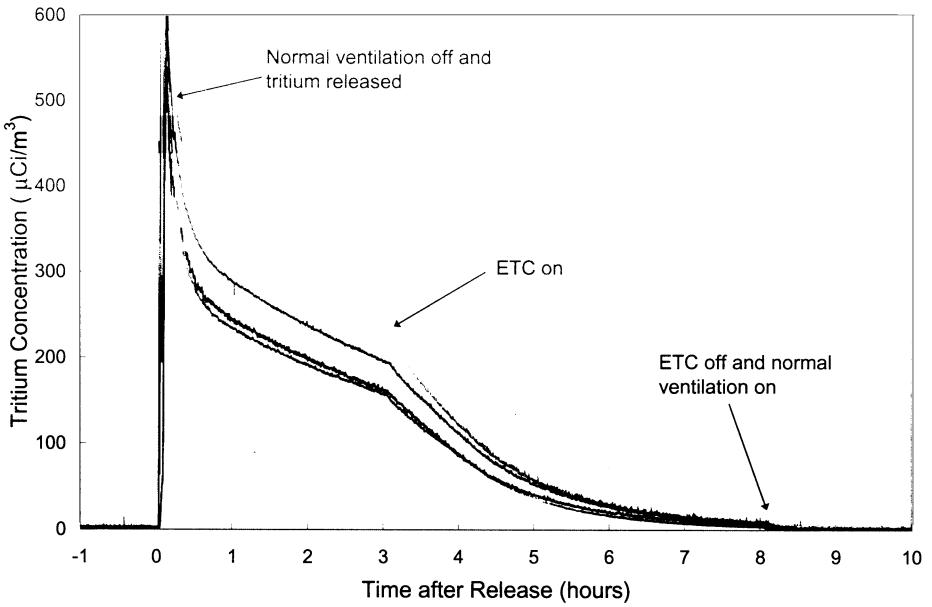

Fig. 6. Tritium concentrations (five ion chambers) vs. time after release for TRE/ETC2.

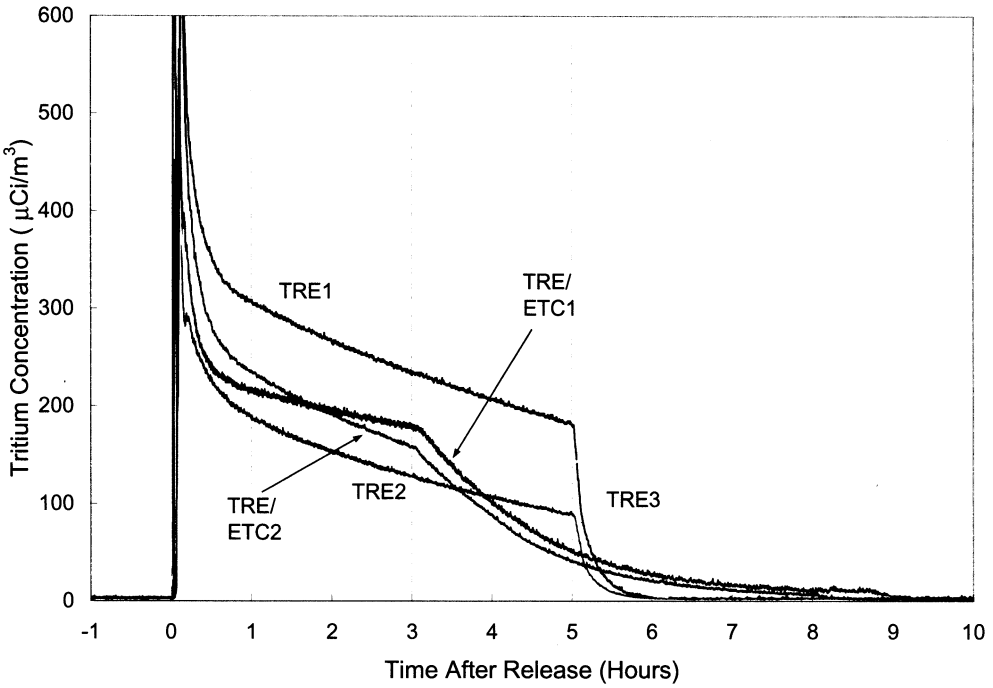

Fig. 7. Room tritium concentration (single ion chamber) for all TRE and TRE/ETC.

show the period of time over which the bubbler sample was collected. The vertical position of the bar shows the HTO concentration indicated by the activity collected in the bubbler. A continuous line was drawn through the centers of the bars to infer the continuous performance.

It is observed that there is a substantial difference between the HTO and total concentration at the beginning. This is explained by the low temperature of the catalyst bed. It appears that once the bed reaches about 420 K that conversion of HT to HTO is complete.

3.4. Surface concentrations

Seven test cell surfaces were swiped immediately before TRE/ETC2. Immediately following the test these same surfaces were swiped again. Then the

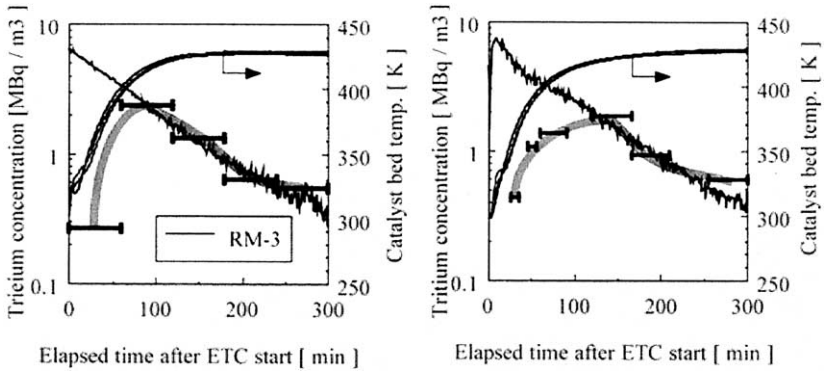

Fig. 8. Total tritium (decreasing line) and HTO (increasing/decreasing line with bars) at catalyst bed exit, and catalyst bed temperature (R.H. axis) for TRE/ETC1 (left plot) and TRE/ETC2 (right plot).

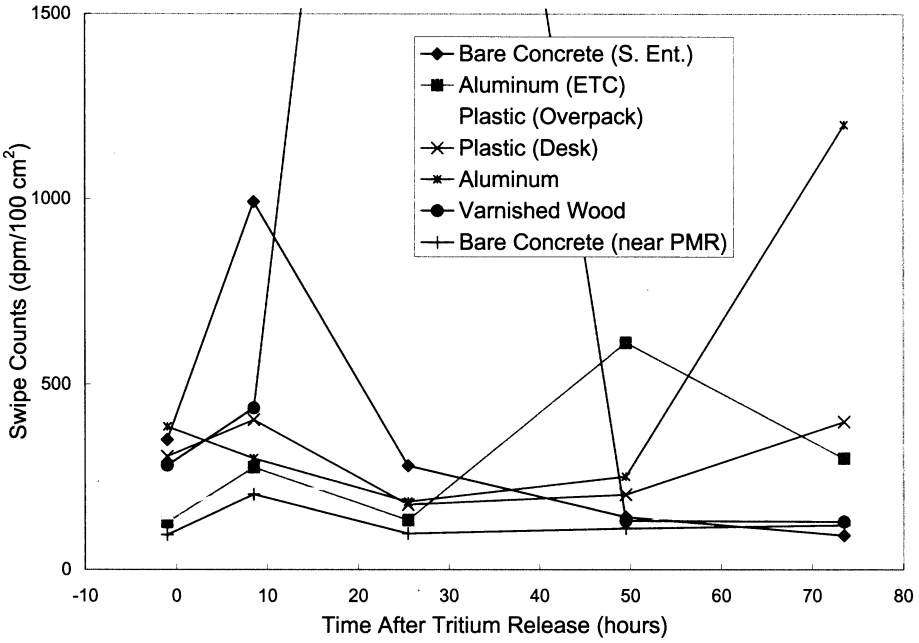

Fig. 9. Tritium surface concentrations on test cell surfaces before and following TRE/ETC2.

surfaces were swiped again on each of three subsequent days. The results are summarized in Fig. 9. For a number of the surfaces, a modest increase in surface tritium is observed immediately following the test (around 50–75%), and these levels drop down to pre-test levels on the following day. However, this effect is very slight. At least three surfaces have larger single-point increases, which appear to have nothing to do with the release experiment. Thus, a 1 Ci release appears to have a short-term, moderate effect on tritium surface concentrations, but this is small compared with routine fluctuations.

3.5. Summary of TRE and TRE/ETC

Table 1 is a summary of results for all TRE and TRE/ETC experiments. A number of useful values result from this study. For the TRE the ventilation was turned off right before the release, the tritium releases were first detected in 23–45 s. For the TRE/ETC1 the ventilation was isolated almost 2 min prior to the release and the time to detection was 56 s. For TRE/ETC2 the ventilation was turned off 21 min prior to release. This may account for the 70 s required to detect the release.

All of the tritium release experiments resulted in room concentrations of about 200 μCi/m^3 at 1 h after the release. This is consistent with the 1 Ci/3000 m^3 = 333 μCi/m^3 combined with the fact that tritium concentrations were naturally decreasing in the room.

In all experiments it appears that it took about 45 min for the room tritium concentrations to become well-mixed.

Using the 18 000 m^3/h stack blower about 30 min was required to reduce the room levels to below one DAC while the 2800 m^3/h required about 260 min.

4. Conclusions

The TRE and TRE/ETC experiments successfully studied the behavior of 1 Ci tritium releases into a practical-scale tritium processing facility. A number of useful factors were drawn from the release data and these have been summarized in this paper.

Table 1
Summary of all TRE and TRE/ETC experiments

Designator	Date	Isolation time (h)	Release time (h)	Ventilation time (h)	Ventilation method	Release location	Release description	Highest concentration ($\mu Ci/m^3$)	Time to detect > DAC (s)	Average concentration at 1 h ($\mu Ci/m^3$)	Time to 'well mixed' (min)	Time to < DAC (min)	Time to bkgd (min)
TRE1	1/29/97	10:29	10:30	15:30	18 000 m^3/h Stack Blower	NE corner of pit	1 Ci	3256	23	225	~45	30	~75
TRE2	12/16/97	10:04	10:05	15:05	18 000 m^3/h Stack Blower	Middle of main floor	1 Ci	4731	45	190	~45	36	~95
TRE3	3/3/98	09:59	10:00	15:12	18 000 m^3/h Stack Blower	Middle of main floor	1 Ci+4 l of D_2	10 351	23	223	~45	29	~85
TRE/ETC1	11/8/00	09:58:30	10:00:20	13:00	2800 m^3/h ETC compressor	Middle of main floor	1 Ci	3330	56	237	~45	263	N/A
TRE/ETC2	3/20/01	08:09	08:30	11:30	2800 m^3/h ETC compressor	Middle of main floor	0.93 Ci	5850	70	188	~45	266	N/A

It was determined that an ETC catalyst bed does not become fully effective for converting HT to HTO until the catalyst reaches about 420 K. The TSTA ETC is not maintained hot at all times and about 2 h was required to reach an effective temperature. This delay must be taken into account when dealing with tritium releases.

The TRE/ETC releases led to only slight increases in tritium surface concentrations. Levels returned to pre-release levels by the following day.

As expected the ETC was much slower in reducing test cell air tritium concentrations compared with normal ventilation. This is because the ETC flow-rate is much lower than that of the stack blower. This factor must be included when considering how tritium releases will be handled.

References

[1] R.V. Carlson, F.A. Damiano, K.E. Binning, Operation of the room air tritium removal system at the Tritium Systems Test Assembly, Fusion Technology 8 (1985) 2190–2195.

[2] F. Sabathier, D. Brennan, N. Skinner, B. Patel, Assessment of the performance of the JET exhaust detritiation system, Fusion Engineering and Design 54 (2001) 547–555.

[3] T. Hayashi, K. Kobayashi, Y. Iwai, T. Yamanishi, M. Nishi, K. Okuno, R.V. Carlson, R.S. Willms, D. Hyatt, B. Roybal, Tritium behavior intentionally released in the radiological controlled room under the US–Japan collaboration at TSTA/LANL, Fusion Technology 34 (1998) 521–525.

[4] T. Hayashi, K. Kobayashi, Y. Iwai, M. Yamada, T. Suzuki, S. O'hira, H. Nakamura, W.M. Shu, T. Yamanishi, Y. Kawamura, K. Isobe, S. Konishi, Tritium behavior in the Caisson, a simulated fusion reactor room, Fusion Engineering and Design 51-52 (2000) 543–548.

[5] Y. Iwai, T. Hayashi, K. Kobayashi, M. Nishi, Simulation study of intentional tritium release experiments in the Caisson assembly for tritium safety at the TPL/JAERI, Fusion Engineering and Design 54 (2001) 523–535.

[6] Y. Iwai, T. Hayashi, T. Yamanishi, K. Kobayashi, M. Nishi, Simulation of tritium behavior after intended tritium release in ventilated room, Journal of Nuclear Science and Technology 38 (2001) 63–75.

[7] K. Kobayashi, T. Hayashi, Y. Iwai, M. Nishi, Results of experimental study on detritiation of atmosphere in large space, Fusion Engineering and Design 58–59 (2001) 1059–1064.