TRITIUM BEHAVIOR INTENTIONALLY RELEASED IN THE RADIOLOGICAL CONTROLLED ROOM UNDER THE US-JAPAN COLLABORATION AT TSTA/LANL

T. Hayashi, K. Kobayashi, Y. Iwai,
T. Yamanishi, M. Nishi, and K. Okuno*

Tritium Engineering Laboratory,
Dept. of Fusion Engineering Research,
Japan Atomic Energy Research Institute,
Tokai-mura, Ibaraki-ken 319-1195, Japan

R.V. Carlson, R.S. Willms,
D. Hyatt, and B. Roybal

Tritium Science and Engineering Group,
Eng. Science and Applications Division,
Los Alamos National Laboratory,
Los Alamos, NM 87545, U.S.A.

ABSTRACT

A series of planned tritium release experiments into the Tritium Systems Test Assembly (TSTA) tritium processing rooms (3000 m^3), were performed under the US-Japan collaboration program at Los Alamos National Laboratory (LANL). These experiments were carried out to acquire data on the behavior of tritium in an actual tritium facility. These experiments were performed safely with no impact on the environment or worker safety using 37 GBq (1 Ci). The results showed that the migration of tritium within the facility was dominated by the residual airflow in the room and reached a uniform value in 30 ~ 40 minutes. After restoring the normal once through ventilation (5 air changes per hour), room tritium levels decreased to background in less than 1 hour. Residual surface contamination was detected (max. 1 Bq/cm^2 level). Residual contamination levels were found to be in order of decreasing contamination: linoleum > epoxy coating > acrylic resin > butyl rubber > stainless steel, from soaking results. The surface contamination increase as a result of the tritium release, was reduced to background with the normal ventilation within a few days.

I. INTRODUCTION

In a DT-fusion reactor, confinement of tritium releases is one of the essential issues for tritium safety of workers and the environment. Tritium gas will be handled under multiple confinements in a fusion reactor and each confinement will have its own detritiation system. This multiple confinement concept has been adopted in worldwide tritium facilities. Safety relevant data has been accumulated on secondary containment systems such as gloveboxes [1~3]. However, only a few reports are available for actual tritium behavior in large containment systems, such as a tritium processing rooms [4]. In order to establish more effective tritium processing safety and confinement systems, actual data of tritium behavior in the large practical multiple containment system should be accumulated in a data base. This database will be useful for comparing with data from small-scale experiments and behavior predicted from system modeling analysis.

To address the above issues a series of intentional tritium release experiments into the radiological controlled areas of TSTA were planned and performed under the US-Japan collaboration program at Los Alamos National Laboratory (LANL). The primary purposes of these tritium release experiments were focused on the following goals. 1) Study tritium mixing and migration within the radiological controlled areas. 2) Monitor airborne tritium removal times from the facility using the normal ventilation until the background tritium levels are reached. 3) Study tritium contamination effects on various surfaces in an operating tritium facility, and surface decontamination by normal ventilation. 4) Measure the conversion of HT to HTO in an operating tritium facility as a function of time.

In this paper, the above experimental results of planned tritium release in the radiological controlled area are summarized and the tritium behavior is discussed. The comparison between the small scale experiments, actual tritium facility releases and computer modeling will be discussed further in a future paper.

II. EXPERIMENTAL

A. Confinement room and apparatus

Figure 1 shows the an artist view of the main tritium handling area of the TSTA in the LANL (solid bold line area). The tritium handling area volume is about 3000 m^3 (~29 x ~12 x ~8.5 m) which has a once through normal ventilation of approximately 5 air exchanges per hour. The tritium area is normally at a negative pressure of approximately 0.2 torr relative to ambient atmospheric pressure. The solid and open circles in figure 1 indicate the

*: Now, Faculty of Science, Shizuoka Univ., Japan.

release points for the tritium releases and the location of the nine ion chamber tritium monitors, respectively. The tritium concentration measured by each tritium monitor was recorded by the Master Data Acquisition and Control system (MDAC) of TSTA. Three water bubbler systems are also installed to measure HTO concentration (see triangle in figure 1).

The 37 GBq of pure tritium for each of the release experiments was filled into the stainless steel container (4.5 ml, 60 torr) on a day before release experiment. At the same time a gas sample was taken. The isotopic composition was determined by mass spectrometer.

In order to measure surface contamination, various materials, such as acrylic resin, butyl rubber, stainless steel (SS316), epoxy coating on SS316, and linoleum were installed at two locations (see squares in figure 1). The contamination was measured both by a smear method and a soaking method. Panels, 50 x 50 cm, of each material and individual samples, 1 x 2 cm, of each material were placed in the tritium areas immediately before each test. A 100 cm^2 area of 50 x 50 cm panels was smeared periodically to determine the tritium contamination. Periodically, one or two of 1 x 2 cm pieces was placed in a vial for soaking with liquid scintillation fluid, following tritium measurement by Liquid Scintillation Counter (LSC).

B. Experimental Method for Planned Tritium Release

First, the sample container was installed to the release system. This system has remotely operated valves (from the TSTA Control Room) and the ability to purge the sample container with N_2. After checking the container and monitors' background, the main cell was locked to prevent personnel entry. The ventilation for the tritium areas was isolated by closing all exhaust and input ventilation valves, stopping the flow of air to or from the facility. Immediately from the control console in the control room, the 37 GBq of tritium gas in the container was released instantaneously and purged by N_2 for a short time to insure the tritium was sweeped from the container. Local tritium concentrations in the cell were monitored by 8-9 ion chambers and 3 water bubblers. Data from the ion chambers was archived every 6 seconds. After 5 hours from the tritium release, normal ventilation system was restored. The tritium in the room was exhausted to the TSTA stack. After the airborne tritium levels decreased to the background, residual contamination on the various surfaces in the tritium areas were surveyed by smear and soaking methods. This sampling was done periodically for more than one week.

The above tritium release experiments were performed at the corner pit and at the center of the main cell on January and December in 1997, respectively.

C. Preliminary Computer Simulation of Tritium Behavior in TSTA

A three dimensional flow calculation code (CFX-F3D), which was available commercially, was used to model the tritium behavior. In order to simplify the actual radiological controlled area, the calculation area was divided to the mesh of about 0.5 x 0.5 x 0.35 m. Most gloveboxes, larger tanks and mezzanine were considered. As shown in the figure 2, each of the six ventilation supply inputs and exhaust outputs were also used to model the actual air flow in the main cell. This current calculation used the air flow due to the ventilation and HTO conversion reaction from elemental tritium using the equation of C(HTO) − k C(tritium gas), considering the experimental tritium concentration [5]. Surface adsorption and desorption reactions can also be used by this code, however, it was not considered in this preliminary calculation. Table 1 shows the input data used in this calculation.

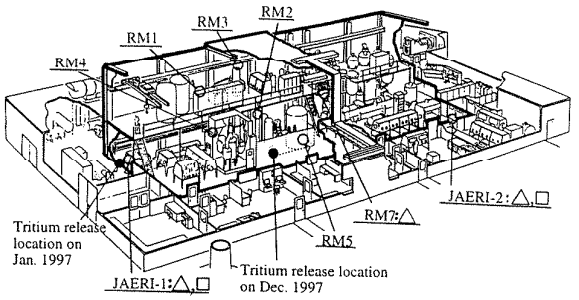

Figure 1 Artist view of TSTA with locations for the tritium release ( ), tritium monitors ( ), bubblers ( ), and clean surface samples( ).

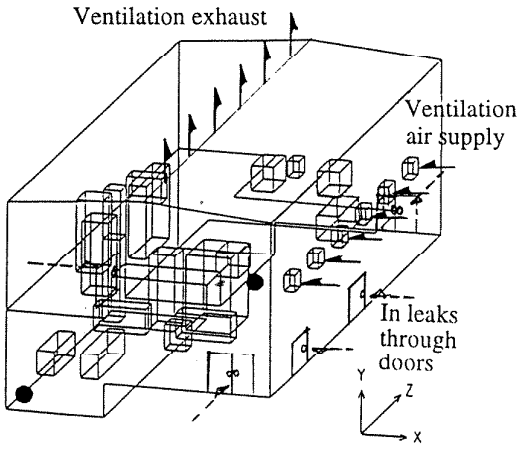

Figure 2 The simplified TSTA main tritium area for three dimensional calculation of tritium behavior

Table 1 Input data for the computer calculation

Species	Diffusion coefficient (m^2/s)	Gravity (kg/m^3)	Viscosity (kg/m/s)
Tritium gas	5.65×10^{-5}		
HTO	2.77×10^{-5}		
Air		1.113	1.85×10^{-5}
Reaction constant (k) :	2×10^{-9} (1/s)		
Room temperature :	293 K constant		
Pressure :	585 Torr (normal LANL atmosphere)		

III. RESULTS AND DISCUSSIONS

The planned release experiments using 37 GBq of pure tritium were performed without any safety problems. During this release experiment, tritium concentration in the non-radiological controlled area, that was surrounded rooms of the main cell, was always almost background.

A. Tritium Mixing, Migration and Removal within the Radiological Controlled Areas

Figure 3 summarizes the local tritium concentration variations after a tritium release from the corner of the tritium area. The result shows that the residual air flow in the cell dominates the migration of tritium. The highest momentary tritium concentration was recorded at approximately 150 MBq/m^3 at nearest monitor to the release point (JAERI-1). All monitors reached approximately a uniform value of 11 ~ 12 MBq/m^3 in 30 ~ 40 minutes. When the release location was changed to the center of the cell, the mixing period was not changed drastically. Although the initial response of each tritium monitors was changed significantly. This fact is consistent with the previously reported behavior at TSTA [4].

After restoring the normal ventilation air input and exhaust flow, the room tritium levels were reduced to the background in less than 1 hour as shown in the figure 4. Because the tritium level of most monitors decreased uniformly, the atmosphere in the cell was mixed completely, even in the actual complex room geometry.

Figure 5 and 6 show typical result of preliminary computer simulation (dashed lines), comparing the above experimental data (solid lines) during the initial mixing and when the normal ventilation was restored, respectively. As shown in the figure 6, simulation result was very nearly identical with the actual tritium behavior during the restoration of normal ventilation. This fact implies that this three dimensional calculation code could be used to evaluate tritium behavior when normal dynamic airflow is restored to the cell. Though the surface contamination of the cell was detectable level as described below, this fraction did not contribute significantly to the airborne tritium removal behavior in this experiment. On the other hand, the tritium behavior during the initial mixing was not consistent with the simulation result, as shown in figure 5. This difference would be caused by the inadequate handling of the residual air flow by the computer code. This is dependent on the mesh size and the simplification of the cell for the calculation. Also, other air currents existed in the cell, even after termination of the ventilation. Some of these are, instrument cooling fans, tritium monitor air pumps, and from air in-leakage from doors, etc. from the non tritium areas which are held at a positive pressure relative to atmospheric pressure. In order to evaluate the actual residual air flow using this simulation code, the case calculations were performed using a 10 times larger residual flow after ventilation isolation or about 15 vol..% /hr of constant air inleakage. The result for RM5 is shown in figure 7, compared with the data of figure 5.

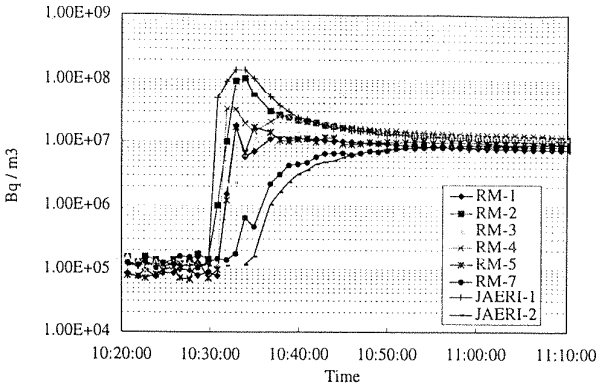

Figure 3 Local tritium concentration for the tritium release experiment from the corner of the main cell (Jan. '97, initial period)

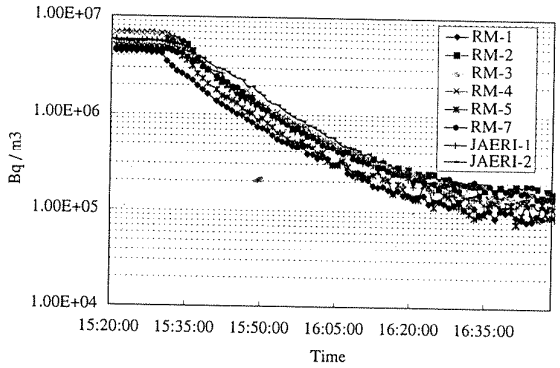

Figure 4 Local tritium concentration at the ventilation (5 times per hour) re-start. (Jan. '97)

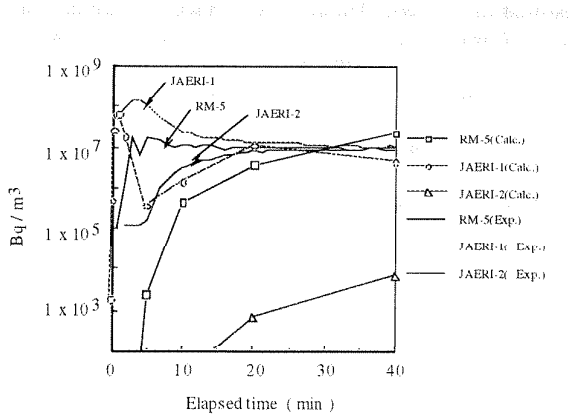

Figure 5 Comparison of measured data (Jan.'97) and calculated value. (initial period of tritium release)

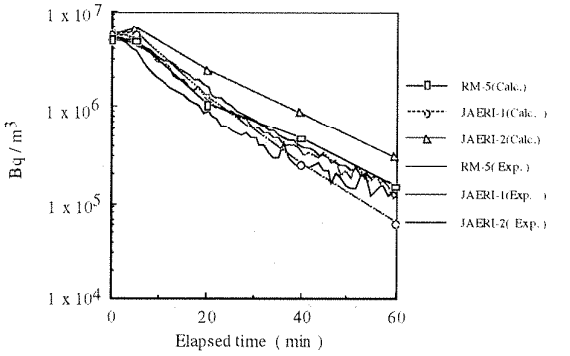

Figure 6 Comparison of measured data (Jan.'97) and calculated value. (after ventilation restored)

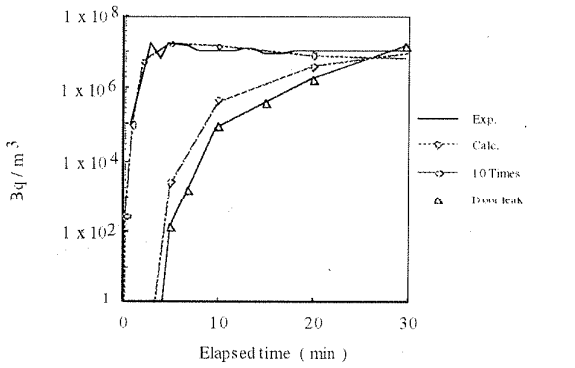

Figure 7 Comparison of measured data (Jan.'97) and calculated value. (after ventilation restored)

This shows that approximately 10 times residual flow could explain the results of this experiment and the preliminary simulation. Constant flow through the door would proceed mixing, however, the effect could not be included in the simulation because of the limitation of the mesh size. In near future, smaller and simple tritium release experiment is planned using the medium caisson in the Tritium Process Laboratory in Japan Atomic Energy Research Institute [6]. The simulation code of tritium behavior in the room would be improved using this TSTA data and the medium caisson one.

B. Tritium Surface Contamination Measurements

Materials for both the smear and soaking method of surface contamination determination were located at two corners of the main cell. Figure 8 shows the typical surface contamination level variations as a function of time after the tritium release. Note the plotted linoleum levels are 1/10 of the actual value. These are shown for the release located at the center of the cell. These were determined with soaking method. Surface contamination levels at a few Bq/cm^2 were detected. These levels were reduced to background after one or two days. The measured contamination levels of each material was roughly the same as determined by either the smear or soaking technique. In order of decreasing contamination as detected by soaking method, the following are the relative contamination levels, linoleum > epoxy coating > acrylic resin > butyl rubber > SS316. This means that some fraction of tritium is absorbed in the bulk of materials and it is difficult to evaluate with the smear technique for each material. Moreover, because the back side of linoleum was a layer structure of fibers, the surface area was larger than that other materials.

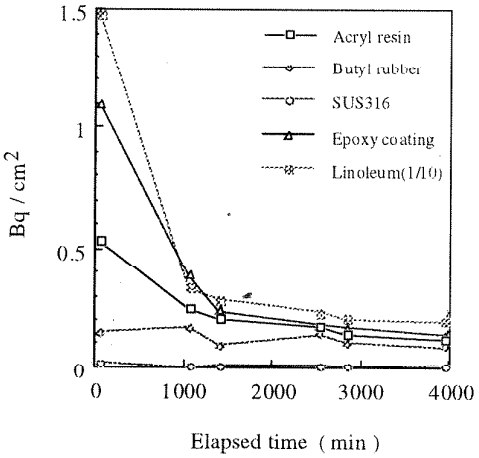

Figure 8 Results of surface contamination for various materials by soaking method.

Thus, the surface contamination of linoleum was much higher than that of other materials as determined by soaking. A significant difference in the surface contamination was not seen for the two sampling locations.

C. HTO Formation Results

HTO concentration in the TSTA tritium area was measured during this experiment and the results are summarized as a function of time in figure 9. Because of the background HTO concentration of the tritium areas of TSTA and the possibility of a small amount of HTO in the released tritium, it was difficult to quantify the conversion rate of HT to tritiated water. However, the conversion rate of tritium gas to tritiated water was determined to be less than 0.5% / day, using the average increase of HTO concentration. The background HTO in TSTA was determined in separate experiments and subtracted from the measured value of HTO during the tritium release. The mean fraction of HTO in the room air was about 1/200 of the total tritium concentration during the release experiment and decreased to the background for a day under normal ventilation.

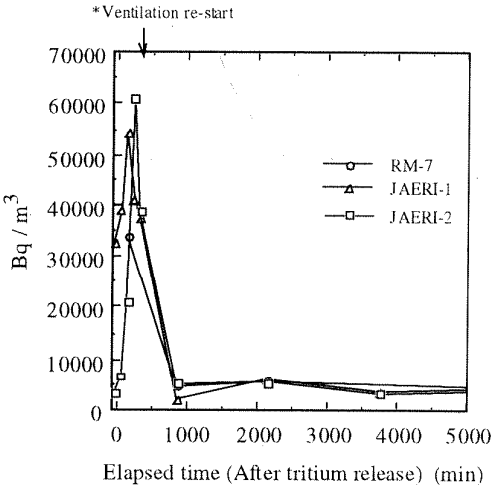

Figure 9 Local HTO concentration in the main cell after the tritium release.

IV. CONCLUSION

A series of planned tritium release experiments at the TSTA/LANL were performed without any impact on worker safety or the environment. The results show that;
1) The migration of tritium within the facility was dominated by the residual airflow in the room and reached a uniform value in 30 ~ 40 minutes. After restoring the normal once through ventilation (5 air changes per hour), room tritium levels decreased to background in less than 1 hour.
2) Residual surface contamination was detected (max. 1 Bq/cm^2 level). Residual contamination levels were found to be in order of decreasing contamination: linoleum > epoxy coating > acrylic resin > butyl rubber > stainless steel, from soaking results. The surface contamination increase as a result of the tritium release, was reduced to background with the normal ventilation within a few days.
3) The maximum conversion rate of HT to HTO was determined to be less than 0.5 % / day.
4) Preliminary calculations using a three dimensional code simulated the tritium behavior in room during normal ventilation, however, the code must be improved to understand the initial tritium behavior just after the tritium release and the isolation of the ventilation.

ACKNOWLEDGMENTS

The authors wish to express their appreciation to Mr. Hirata, Mr. Shibuya and Mr. Kato (JGC Corporation) for their help of three dimensional analysis of tritium behavior. The authors also gratefully acknowledge the US-Japan collaboration project for supporting this experiments.

REFERENCES

[1] Final Safety Analysis Report of Tritium Systems Test Assembly at the Los Alamos National Laboratory, TSTA-SAR, Los Alamos National Laboratory (1996).
[2] Y. Naruse, Y. Matsuda and K. Tanaka, " Tritium Process Laboratory at the JAERI ", *Fusion Engineering and Design*, 12, 293 (1990).
[3] P. Schira, E. Hutter, G. Jourdan and R.-D. Penzhone, " The Tritium Laboratory Karlsruhe; Laboratory Design and Equipment ", *Fusion Engineering and Design*, 18, 19 (1991).
[4] J.R. Bartlit, J.L. Anderson, R.A. Jalbert, R.V. Carlson, K. Okuno, T. Ide, H. Fukui, M. Enoeda and Y. Naruse, " Dispersion and Removal of Tritium Released into the Main Cell of TSTA", *Proc. of 13th SOFE, Knoxville, TN.,U.S.A.*, 798 (1989).
[5] H. Noguchi, C. Easterly and M.R. Bennett, " Conversion of Low Concentration Tritium Gas to Tritiated Water ", *Fusion Technology*, 16, 137 (1989).
[6] S. O'hira, T. Hayashi and K. Okuno, " Tritium Safety Related Studies at TPL of JAERI", *Fusion Energy*, 16, 219 (1998).