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TRITIUM SURFACE CONTAMINATION

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ABSTRACT

Glovebox wipe surveys were conducted to correlate surface tritium contamination with atmospheric tritium levels. Surface contamination was examined as a function of tritium concentration and of tritium form, HT/T₂ and HTO. The relationship between atmospheric HTO concentration and cleanup time was also investigated.

INTRODUCTION

The next generation of fusion devices will probably utilize a deuterium-tritium-lithium fuel cycle. As a result, tritium handling systems have been incorporated into these designs to manage tritium in all aspects of operation, including the control and recovery of tritium in buildings. It has been shown that tritium contamination in the reactor hall environment can have a significant impact on the total dose received by the worker as well as on the capital and operating costs of systems required to maintain a given atmospheric tritium level. The maximum atmospheric concentration of HTO that can be tolerated with minimal worker protection and still maintain the dose <5 rem/yr is 5 $\mu\text{Ci}/\text{m}^3$. Higher concentrations could be tolerated with protective clothing; i.e., air-supplied bubble suits.

During steady-state operation of a fusion reactor, a steady leakage of tritium into the reactor hall is expected. Because it is desirable to be able to enter the reactor hall as soon as possible after the reactor has been shut down, atmospheric tritium levels need to be reduced in a minimum amount of time. If it is assumed that the reactor hall atmosphere cannot be vented to the atmosphere and, in the ideal case, that the reactor hall atmosphere is uniformly mixed, that there are no tritium leaks while the reactor is shut down, and that there is no outgassing from components in the reactor hall, the time to reduce atmospheric tritium concentration to prescribed levels can be easily calculated. Making these assumptions, the average atmospheric residence time, t_r , can be calculated according to the expression:

$$t_r = V/q = 100\%/F \quad (1)$$

where V = volume of reactor hall (or glovebox)
 q = flow rate (to a tritium recovery system)
 F = Vol % change in atmospheres/min.

In this case, the atmospheric tritium concentration at any time, t , is given by the expression:

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$$C = C_0 \exp(-t/t_r) \quad (2)$$

where C_0 = the initial concentration.

In practice, however, tritium diffuses into components during operation and diffuses back into the containment atmosphere after shutdown, lengthening the time necessary to reduce atmospheric tritium contamination to acceptable levels. If the reactor hall materials outgas at a relatively high rate, mathematical models indicate that the walls could become a significant tritium source during decontamination efforts.

In order to obtain a further understanding of this phenomenon, a study was undertaken to correlate glovebox atmospheric tritium concentrations with surface contamination in an effort to predict the degree of contamination in the reactor hall of a fusion plant. Surveys inside gloveboxes were made to determine surface activity levels on stainless steel after exposure to tritium over a range of concentrations and exposure times. In addition, the relationship between atmospheric concentration and cleanup time was also investigated.

RESULTS

To assess the magnitude of tritium contamination on glovebox surfaces, several experiments were performed. Wipe surveys were taken on stainless steel surfaces that had been exposed to a wide range of tritium concentrations in both the elemental and oxide forms. Atmospheres in the glovebox were either air or nitrogen, though boxline oxygen and humidity were not monitored.

In all experiments, sampled areas were estimated to be approximately 100 cm². Surface wipes were taken using filter paper discs, and the degree of contamination from the glovebox atmosphere was determined by exposing 1 to 3 filter papers for a period of about 1 min. The

contribution from ambient glovebox atmospheric tritium was subtracted from apparent wipe counts to establish the extent of actual surface contamination. All wipes were counted in a conventional Nuclear Measurements Corporation PC-4 gas proportional counter.

The data in Table 1 represent two sets of experiments used to assess the degree of HTO contamination on stainless steel surfaces and its contribution to the cleanup time required to attain a steady state atmospheric tritium level. During one set of experiments, Tests 5-9, the glovebox was maintained in a static condition. In the other experiment, atmospheric tritium concentration and surface contamination were measured as a function of time to determine the effect of surface contamination on the overall removal rate. Best fit correlations using regression techniques have been performed on the data. The relationship between atmospheric concentration and surface contamination is shown in Figure 1. The relationship was found to be:

$$y = 1.11 \exp(4.6x) \quad (3)$$

where x = atmospheric HTO concentration, mCi/m³
 y = surface level contamination,
 dpm x 10⁻³/100 cm²

Tests 10-16 from Table 1 established the relationship between the atmospheric concentration x and the cleanup time, t , (hours). This relationship is given by:

$$x = 1/(1.2 + 0.035 t) \quad (4)$$

In this experiment, $t_r = 800$ min, and $F = 0.13\%$. Based on these assumptions with the initial atmospheric tritium concentration, C_0 , of 0.81 mCi/m³ and $t_r = 800$ min, the atmospheric concentration, C , as a function of time can be calculated from Eq. (2). In the ideal situation, as shown in Figure 2, the time to reduce atmospheric tritium concentration is

Table I - SURFACE TRITIUM CONTAMINATION ON STAINLESS STEEL GLOVEBOX
AS A FUNCTION OF HTD CONCENTRATION^a

Test Order	HTD Concentration (mCi/m ³)	Time at This HTD Concentration (hr)	Average Surface Level ^b (dpm x 10 ⁻³ /100cm ²)	Average Background ^c (dpm x 10 ⁻³ /100cm ²)
1	0.090	<336	1.05	0.9
2	0.260	36	4.8	9.6
3	0.390	36	7.5	23.1
4	0.810	48	49.5	13.2
5	0.625	168	16.8	25.2
6	0.600	48	18.9	27.3
7	0.600	48	17.4	16.2
8	0.635	96	13.8	16.5
9	0.625	48	16.2	20.2
10	0.810 ^d	24	43.5	18.6
11	0.490	24	18.0	12.9
12	0.300	24	3.6	11.7
13	0.255	24	2.64	7.5
14	0.230	24	2.67	5.7
15	0.200	24	4.8	3.3
16	0.185	<24	2.88	2.7

^a Air purged glovebox, 5 l/min, except during Tests 5-9.

^b Sampled areas were 100 cm². The level is the average of 9 to 12 locations in the glovebox with background subtracted.

^c The background is the average of at least two samples exposed to the atmosphere for 1-3 min intervals.

^d A 5-day interval existed between Tests 9 and 10.

dramatically reduced. For example, the time required to reduce tritium levels from 0.810 mCi/m³ to 5 μ Ci/m³, a level that would allow manned access to a reactor hall with minimal worker protection, is about 3 days. The actual data, however, indicate the time required to reduce levels to 5 μ Ci/m³ could be several months in cases where tritium extensively soaks into the walls and then permeates back into the containment atmosphere, as evidenced in this experiment. Even with initial concentrations of 50 μ Ci/m³ and flow rates of 0.13 vol %/min, several weeks could be required in this case to attain 5 μ Ci/m³, a lengthy downtime in the case of a commercial fusion reactor.

The data in Table 2 represent the experiments used to assess the degree of HT/T₂ contamination on a new stainless steel glovebox surface. Atmospheric tritium concentration and surface contamination were measured to determine the relationship, if any, between the two. In these experiments, the time variable could not be controlled due to the operations that were currently being conducted in the box. The relationship between atmospheric concentration and surface contamination (shown in Figure 3), was found to be:

$$y = 2.54x^{1.03} \quad (5)$$

Figure 1 - Atmospheric concentration in relation to wipe results.

where x = atmospheric HT/ T_2 concentration,

$$\text{mCi/m}^3$$

y = surface level contamination,

$$\text{dpm} \times 10^{-3}/100 \text{ cm}^2$$

The relationship between atmospheric concentration and cleanup time was further examined during the decommissioning of three tritium handling gloveboxes. During the period of this study, the boxline purge system was configured to provide a single pass sweep through the boxline by the purge gas. (See Figures 4a,b,c.) This method was chosen because the tritium concentrations in the atmosphere in box 3 were substantially higher than those in box 2, which, in turn, were greater than levels in box 1. (See Table 3.) In addition, due to the high tritium concentrations, high purge rates were used, resulting in low residence times, t_r , and high vol %/min changes, F . When atmospheric tritium concentration as a function of time was plotted and fitted to a curve by regression

techniques, the equation describing the data was of the same form as that observed previously, in particular $x = 1/(A + Bt)$. The relationship between atmospheric concentration, x , in mCi/m^3 and cleanup time, t , in hours for box 1 is given by:

$$x = 1/(0.69 + 0.017t) \quad (6)$$

This equation has been plotted in Figure 5.

Once atmospheric levels in box 1 reached a level that allowed removal of hood fronts, the pass door was sealed, the purge was configured so that box 1 was isolated, and the purge was instituted in box 2. (See Figure 4b.)

Data obtained from purging box 2 yielded similar results. A best fit curve of the data yielded the following equation relating atmospheric concentration and time:

Figure 2 - Atmospheric concentration as a function of time.

$$x = 1/(0.14 + 0.013t) \quad (7)$$

This has been plotted in Figure 6.

Two sets of data were generated during the atmospheric decontamination of box 3. (See Tables 3 and 4.) During the rearout of the fumehood under box 3, it was discovered that two water transfer lines containing a few cubic centimeters of 750 Ci/l water were now open to the boxline. After these were removed, a significant reduction was observed in atmospheric contamination levels. Before removing these lines, the equation describing the atmospheric concentration at any time, t , was:

$$x = 1/(0.0084 + 1.6 \times 10^{-4}t) \quad (8)$$

Once this source term had been removed, atmospheric levels decreased substantially, yielding the equation:

$$x = 1/(0.046 + 0.0049t) \quad (9)$$

These data are plotted in Figures 7 and 8.

It should be noted that in these instances it was consistently found that atmospheric contamination was best described by the equation:

$$x = 1/(A + Bt) \quad (10)$$

At the initiation of decontamination, $t = 0$, and $x = C_0$, the initial glovebox atmospheric

Table 2 - SURFACE TRITIUM CONTAMINATION OF STAINLESS STEEL GLOVEBOX
AS A FUNCTION OF HT/T₂ CONCENTRATION^a

Test Order	HT/T ₂ Concentration (mCi/m ³)	Average Surface Level ^b (dpm × 10 ⁻³ /100cm ²)	Average Background ^c (dpm × 10 ⁻³ /100cm ²)
1	0.40	0.39	0.99
2	0.80	3.80	0.48
3	0.60	1.02	1.36
4	0.18	0.25	0.51
5	0.08	0.63	1.16
6	0.12	0.39	4.55
7	0.70	0.86	35.6
8	1.35	6.23	6.62
9	1.65	3.49	10.69
10	1.75	5.60	11.46
11	0.30	3.85	32.75
12	27.00	18.60	74.17

^aNitrogen purged glovebox.

^bSamples areas were 100 cm². The level is the average of 6-8 individual locations with background subtracted.

^cThe background is the average of at least 2 samples exposed to the atmosphere for a short interval.

Figure 3 - Atmospheric concentration (HT) in relation to wipe result.

a. 3 box purge configuration.

b. 2 box purge configuration after glovebox 1 hoodfronts removed.

c. Single box purge configuration after glovebox 1 and 2 hoodfronts removed.

Figure 4 - Glovebox purge configurations for atmospheric decontamination.

Figure 5 - Glovebox 1 atmospheric decontamination.

Figure 6 - Glovebox 2 atmospheric decontamination.

Table 3 - VARIATION OF ATMOSPHERIC CONCENTRATION WITH TIME^a

Time (hr)	Atmospheric Concentration (mCi/m ³)		
	Box 1	Box 2	Box 3
0	3.0	6.0	130.0
6	1.3	4.0	96.0
24	1.0	2.0	85.0
48	0.6	1.3	60.0
72	0.5	1.2	50.0
96	0.375	0.65	28.0
120	0.230	0.40	9.1
192	0.330	0.36	8.15
216	0.23	0.34	NA ^b

^at_r = 88 min.

^bNA - Equipment failure made data unavailable.

Table 4 - VARIATION OF ATMOSPHERIC CONCENTRATION WITH TIME, GB 3 WATER SOURCE TERM REMOVED^a

Time (hr)	Atmospheric Concentration (mCi/m ³)
0	18.0 ^b
48	2.9
72	1.95
120	1.60
144	1.85
264	1.25
288	0.50

^at_r = 88 min.

^bAtmospheric equilibrium value after water source removed.

tritium concentration. Hence, $A = 1/C_0$. It is theorized that B is a function of V, the glovebox (or reactor hall) volume, q, the purge rate, and C₀ the initial atmospheric level. Based on this assumption and given the physical parameters for the various glovebox configurations, B can be calculated.

$$B = 1/(C_0 t_r) \quad (11)$$

This satisfies the form of the equation $1/(A + Bt)$ dimensionally and was therefore examined as a possible means for predicting the coefficient B. However, when this was done, it was found that the calculated value of the

Figure 7 - Glovebox 3 atmospheric decontamination, source term present.

Figure 8 - Glovebox 3 atmospheric decontamination, source term removed.

constant B differed from that obtained when regression techniques were used to fit the data collected during the tests. When the actual value for the constant B was compared to the calculated value, a relationship was discovered indicating that:

$$B_a = 0.30B^{1.36} \quad (12)$$

where B = calculated value of the coefficient

B_a = actual value of the coefficient B

This relationship is plotted in Figure 9. As can be seen in the figure, there is a great deal of variation between the data and the best fit curve.

CONCLUSIONS

Wipe surveys taken in HTO handling gloveboxes indicate that the degree of surface contamination varies with the atmospheric concentration according to the expression:

$$y = A \exp(Bx) \quad (13)$$

Atmospheric decontamination studies on several HTO tritium handling gloveboxes indicate that decontamination may be best represented by an expression of the form given by Eq. (10). When compared to the ideal case, the time requirement for detritiation using comparable purge flow rates is quite lengthy. This implies that the required detritiation system must be capable of handling a large volumetric flow rate or that alternatives must be pursued that would minimize tritium soaking into the reactor hall walls. When tritium soaking of the walls is considered, capital costs may escalate to the point that this effect will have a major impact on the cost of the detritiation system used. The alternative would be to minimize tritium soaking by maintaining reactor hall HTO levels near the $5 \mu\text{Ci}/\text{m}^3$ range during normal operations. This could be accomplished by eliminating or minimizing tritium releases to the

Figure 9 - B predicted versus B actual.

reactor hall while maintaining normal reactor hall ventilation, or by constantly treating the atmosphere in a decontamination system.

While it is easy to determine the constant A in Equation 10, B appears to be a function of several variables, including the initial concentration, the purge flow rate, and the volume of the enclosure. These variables are easily measured, satisfy the relationship dimensionally, and were therefore examined as a means to predict this coefficient. The data in Tables 3 and 4 obtained during the decontamination of Box J suggest that B does vary with the initial concentration, C_0 , when V and q are held constant. However, when B is calculated using the predicted model based on these parameters, it was found that the calculated value varied from that resulting from curve fitting using regression techniques. This variance could be the result of some other factor that is not as easily quantified. Since it appears that the relationship between the observed value of B as determined by regression techniques and that

calculated based on the system parameters fits an expression of the form $y = Ax^B$, this factor may arise from the type of surface, the surface area, or some interaction of measurable variables that is dimensionless. However, due to the limited data available, the significance of the relationship between the calculated value of B and that determined by regression methods is uncertain. Only by further study will it be possible to determine what this factor is.

Surveys in HT/T₂ handling gloveboxes indicate that surface contamination varies with atmospheric concentration according to the expression:

$$y = Ax^B \quad (14)$$

Because these experiments were conducted in a new glovebox (i.e., its first exposure to tritium), it was expected that the best fit curve would pass through the origin. Because this was not the case, it is possible that some soaking of the surfaces had already occurred,

even though their exposure time was relatively short. In addition, it can be seen that the form of the best fit equation is different from the case of a surface exposed to HT0.

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