

**SIMULATION OF LARGE SCALE AIR DETRITIATION OPERATIONS BY  
COMPUTER MODELING AND BENCH-SCALE EXPERIMENTATION\***

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Summary

Although some experience has been gained in the design and construction of 0.5 to 5 m<sup>3</sup>/s air-detrition systems, little information is available on the performance of these systems under realistic conditions. Recently completed studies at ANL have attempted to provide some perspective on this subject. A time-dependent computer model was developed to study the effects of various reaction and soaking mechanisms that could occur in a typically-sized fusion reactor building (~10<sup>5</sup> m<sup>3</sup>) following a range of tritium releases (2 to 200 g). In parallel with the computer study, a small (~50 liter) test chamber was set up to investigate cleanup characteristics under conditions which could also be simulated with the computer code. Whereas results of computer analyses indicated that only ~10<sup>-3</sup>% of the tritium released to an ambient enclosure should be converted to tritiated water, the bench-scale experiments gave evidence of conversions to water greater than 1%. Furthermore, although the amounts (both calculated and observed) of soaked-in tritium are usually only a very small fraction of the total tritium release, the soaked tritium is significant, in that its continuous return to the enclosure extends the cleanup time beyond the predicted value in the absence of any soaking mechanisms.

Introduction

The need for large capacity air handling and detrition systems to service ambient operated fusion buildings is well recognized.<sup>1-3</sup> The cost of such systems should scale roughly linearly with the volumetric air flow rate which is in turn determined by (1) the permissible length of time to complete a cleanup operation following the largest conceivable tritium release to a given enclosure and (2) the overall volume of that enclosure. Recent fusion reactor design and safety analyses<sup>1-3</sup> have uncovered at least two dominant incentives for achieving the shortest possible cleanup times. Firstly, short cleanup times would permit timely access to the reactor building when manual maintenance or repair is required, thereby reducing reactor downtime in most cases. Secondly, the amount of tritium soaked onto and into the surfaces in the reactor building is expected to increase with increasing cleanup duration and, as a consequence, the subsequent release of this "soaked" tritium could further extend the time period required to reduce the atmospheric tritium concentration to breathable levels.

Some experience has been gained in the design and construction of 0.5 to 5 m<sup>3</sup>/s air detrition systems.<sup>4-7</sup> What has not been investigated in any detail is the actual performance of fully integrated cleanup systems during massive detrition operations on large enclosures. In particular the impact of various chemical reaction and soaking mechanisms on the progress of the cleanup operation is not well understood. A series of studies to evaluate some of these factors was carried out at ANL during 1977.

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The approach taken in these studies involved (1) the development of a time dependent computer model to simulate anticipated physical and chemical responses in typical enclosures following a large tritium release and (2) a series of bench-scale tests with tritium in a relatively small enclosure under conditions that were intended to emulate a reactor building but on a much smaller scale. The following sections of this paper summarize in order (1) the computational model for cleanup analyses, (2) the results of computer studies of cleanup operations on conceptual fusion reactor buildings, (3) the methodology for and results of bench-scale tests with the small enclosure, and (4) attempts to analyze the bench-scale data using the computer model.

Description of the Computer Model

This section summarizes the important features of the model developed to simulate hypothetical air-detrition scenarios under conditions anticipated in near-term fusion reactor containment buildings. The logic of the resulting code (TSOAK) is based largely on (1) correlations of existing experimental data for pertinent chemical and physical processes (involving tritiated species) with the type of phenomena likely to occur in a large enclosure, (2) derivation of a set of time dependent simultaneous equations to describe the cleanup operation, and (3) selection of an efficient algorithm to solve the resulting set of equations. A detailed description of the model is given in reference 8. In brief, the steps built into the model are as follows:

Cleanup Operation

The principal pathway for removal of tritium bearing species from a reactor building atmosphere following an accidental release is the cleanup operation itself. In the model it is assumed that the air in a room of volume V is circulated through a detrition system at a volumetric flow rate Q and that the efficiency of the detrition system for removal of tritium-containing compounds in the air (regardless of their chemical form) is ε. The rate of reduction in the concentration of tritiated species, N<sub>T</sub>, with time, t, by this mechanism is, therefore, given by

$$\left(\frac{\partial N_T}{\partial t}\right)_A = -\left(\frac{\epsilon Q}{V}\right) N_T = -AN_T \quad (1)$$

$$\text{i.e., } A = \frac{\epsilon Q}{V} \quad (2)$$

Reaction of T<sub>2</sub> with H<sub>2</sub>O and O<sub>2</sub>

There is ample evidence<sup>2,8</sup> to support the assumption that T<sub>2</sub> released to an atmosphere containing an appreciable amount of oxygen and moisture will react with the O<sub>2</sub> and H<sub>2</sub>O to form HTO, T<sub>2</sub>O, and HT.

Accordingly, a term to accommodate the production of tritiated water is included in the model. The basis for the development of this term is described in reference 8.

#### Permeation of T<sub>2</sub> and HT into Building Surfaces

The permeation of molecular tritium species (T<sub>2</sub> and HT) into building surfaces is dealt with in an approximate manner by summing the permeabilities of all structural surfaces based on an assumed steady-state condition<sup>8</sup> where the driving force is proportional to the half-power of the T<sub>2</sub> and HT concentrations in the building atmosphere.

#### Absorption and Release of HTO and T<sub>2</sub>O

The adsorption and release of tritiated water (referred to hereafter as TO) also had to be treated in a highly approximate manner. The rate of adsorption of TO was taken to be proportional to the concentration of TO in the enclosure, and the corresponding proportionality constant was determined by summing the estimated adsorptivities of the different surfaces in the enclosure. A second proportionality constant is included in this part of the model<sup>8</sup> to correlate the rate of release of adsorbed tritiated water with the total amount adsorbed on the building surfaces at any given time during cleanup.

#### Soak Effect and Amount Soaked

The soak effect is defined as the difference between the amount of tritium in the enclosure under ideal cleanup conditions, i.e., those strictly obeying equation (1), and the amount calculated by TSOAK when reaction and soaking mechanisms are active. The soak effect and the amount of soaked tritium are determined in a stepwise fashion<sup>8</sup> along with the rest of the integration required to determine ambient concentrations of T<sub>2</sub>, HTO, and HT.

#### Application of the Soaking Code (TSOAK) to a Typical Reactor Building

In order to evaluate the application of TSOAK to a typical reactor building, a set of representative parameters were developed based on current projections for near-term tokamak reactors.<sup>2,3</sup> Results of soaking calculations for a "base case" scenario involving the cleanup of tritium releases in the range 2 to 200 g to a 1.6 x 10<sup>5</sup> m<sup>3</sup> room by a 5 m<sup>3</sup>/s air detritiation are presented in Table 1. Also shown in Table 1 are the results obtained by varying selected "base case" parameters. As indicated in the table, the amount soaked and the cleanup time are most sensitive to air flow rate. However, inclusion of 20 m<sup>2</sup> of cold surface, which is simulated by a sticking probability near unity coupled with a somewhat higher long term release coefficient (+20 m<sup>2</sup> at <0°C in Table 1), led to the prediction of a considerably extended cleanup time due to the slow back-release of condensed HTO.

Graphical computer output for the "base case" and the case involving the simulated cold surface are given in Figs. 1 and 2. The two solid curves in Fig. 2a correspond to the ideal cleanup response (lower plateauing curve) and the TSOAK calculated response for the base case (upper plateauing curve). In Fig. 1a both the ideal and TSOAK curves are identical and, hence, overlapping. The plateauing of the cleanup curves (solid lines) in Figs. 1a and 2a results from the inclusion of a 4 μCi/m<sup>3</sup> asymptote to more realistically represent the kinds of terminating effects (due to background

signals) that may be encountered in actual monitoring operations. The need for this asymptote is more clearly evident from the results reported in the next two sections. The abrupt termination in the "soak effect" and "amount soaked" curves in Fig. 2a is a result of oscillations that develop in the differential equation solver when the amount of soaked tritium is large.

#### Bench-Scale Tests of Air Detritiation

In order to provide a test of the computer model for air detritiation analysis, a bench-scale experiment was set up to simulate, at least in part, the conditions anticipated for larger enclosures following a tritium release. The experimental assembly consisted of an ~50-liter cubical plastic desiccating box with a flat plexiglass door clamped to one face of the box and sealed by a circumferential rubber gasket. This box (hereafter referred to as the enclosure) was attached to two separate gas circulation loops. One loop (monitoring loop) was connected directly to a flow-through type tritium monitor (Iriton Model 955B) having its own internal circulation pump. The other loop (processing loop) consisted of a flow meter, an oxidizer bed (Hopcalite) operated at ~550°C, a moisture trapping bed (4A molecular sieve) operated on 0°C, and a diaphragm-type circulation pump. Typical experiments were carried out by (1) injecting enough tritium as HT to raise the tritium level in the enclosure to from 1000 to 3000 μCi/m<sup>3</sup>, (2) circulating the enclosure atmosphere through the monitoring loop until a stable tritium signal was achieved, and then (3) starting up circulation through the processing loop.

To a first order of approximation, a cleanup operation carried out in the manner described above should proceed with a simple exponential dependence on time as given by equation (1). In actual practice, the uncertainties in measuring flow rate and enclosure volume (at best a few percent in each case), coupled with the fact that the efficiency is near unity, make it difficult to determine the extent to which equation (1) is obeyed. This is important because, when the trapping efficiency is only slightly less than unity, a significant amount of tritiated moisture can be transported back to the enclosure. The consequences of these uncertainties should be borne in mind when considering the results described below.

Figure 3 shows the results of typical detritiation simulations on the enclosure. The apparent obviance of equation (1) persists from the starting point (t = 0, N<sub>T</sub> = 1300 μCi/m<sup>3</sup>) to an N<sub>T</sub> value roughly 40 times lower, after which a pronounced tailing of each cleanup curve is observed. In order to perform the experiments shown in Fig. 3, an electrical feed-through was adapted to the enclosure to permit operation (inside the enclosure) of a circulation fan and a calor heated stainless steel plate with ~200 cm<sup>2</sup> of surface area. As indicated in Fig. 3, the curves therein represent results of cleanup operations performed on the enclosure for various sets of operating conditions involving the fan and heated plate. Operation of the fan during detritiation (Case B) seems to enhance the tailing effect, possibly by accelerating the rate of the reaction/adsorption/release mechanisms involving HT and, more importantly, HTO. Operation of the hot plate at ~200°C together with the fan (Case C) demonstrated only moderately different detritiation characteristics relative to Case B. Although the hot plate might be expected to cause an increase in the rate of HTO formation, it also raises the enclosure atmosphere temperature by ~20°C, which would tend to reduce the amount of adsorbed HTO.

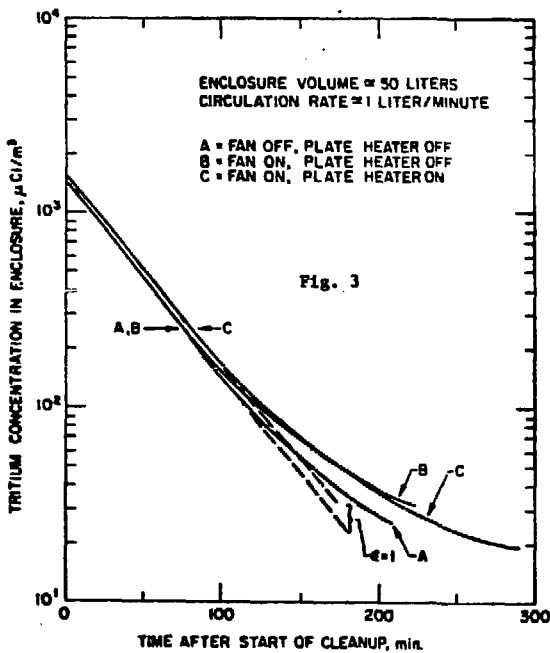
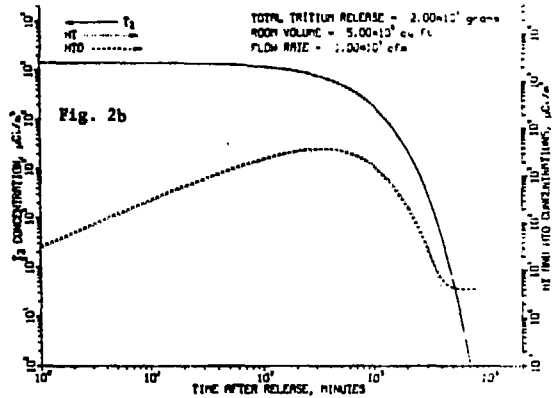
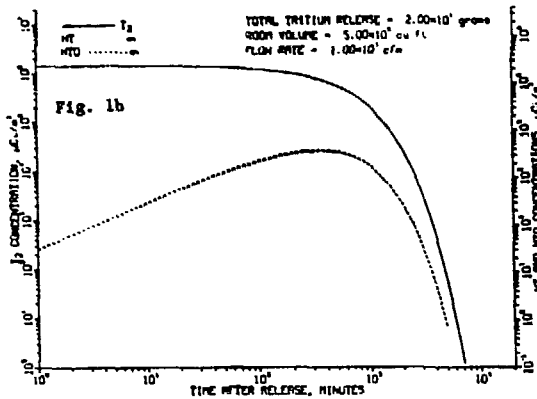
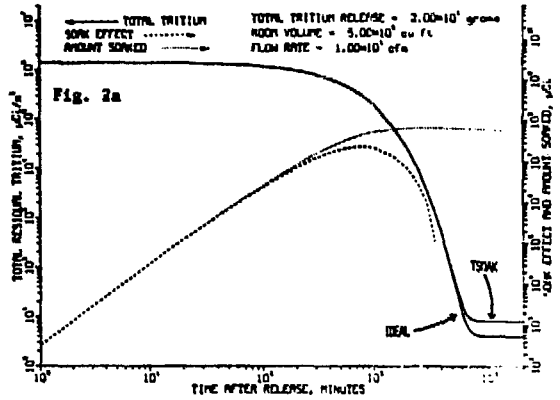
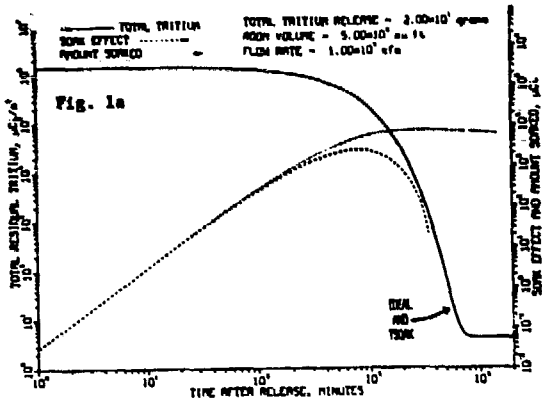


Fig. 1. Graphical representation of TSOAK calculations for a "base case" scenario of a 20-g  $I_2$  release (see Table I and text). The total tritium concentration, soak effect, and amount soaked as functions of time are given in Fig. 1a. The concentrations of  $I_2$ , HTO, and HT as functions of time are given in Fig. 1b.

Fig. 2. Graphical representation of TSOAK calculations for the "base case" scenario of a 20-g  $I_2$  release, modified to include 20 m<sup>2</sup> of condensing surface (i.e., at  $\leq 0^\circ\text{C}$ ). The total tritium concentration, soak effect, and amount soaked as functions of time are given in Fig. 2a. The concentration of  $I_2$ , HTO, and HT as functions of time are given in Fig. 2b.

Fig. 3. Observed cleanup curves (solid lines) for selected experiments with the bench-scale air decontamination apparatus. Enclosure conditions are indicated on the figure. Also shown are ideal cleanup curves for cases A and B (lower dashed curve) and case C (upper dashed curve.)

**Table 1**  
**SELECTED RESULTS OF AIR CLEANUP CALCULATIONS USING TSOAK**

**Base Case Parameters:**

Building Volume	= $1.6 \times 10^5 \text{ m}^3$	Reaction Rate	= $1.5 \times 10^{-11} \text{ m}^3/\mu\text{Ci}\cdot\text{min}$
Process Efficiency	= 1.0 (100%)	HTO Adsorption Rate	= $1.0 \times 10^{-7}/\text{m}^2\cdot\text{min}$
Air Flow Rate	= $5 \text{ m}^3/\text{s}$	HTO Release Rate	= $1.0 \times 10^{-6}/\text{min}$
Building Surface Area	= $2 \times 10^6 \text{ m}^2$	BPT <sup>a</sup>	= $0 \text{ }\mu\text{Ci}/\text{m}^3$

Conditions	Release Size (g)	Amount Soaked (Ci)	Cleanup Time (hr)
Base Case	200	$6.3 \times 10^{-1}$	142
	20	$6.8 \times 10^{-2}$	125
	2	$7.0 \times 10^{-5}$	100
Base Case + Increase Air Flow Rate by 10-X	200	$7.6 \times 10^{-3}$	14
	20	$7.7 \times 10^{-5}$	12
	2	$7.7 \times 10^{-7}$	10
Base Case + Increase Reaction Rate by 100-X	200	$6.0 \times 10^0$	142
	20	$3.6 \times 10^{-1}$	125
	2	$6.3 \times 10^{-3}$	100
Base Case + Increase Adsorption Rate by 100-X	200	$<5.0 \times 10^1$	Tailing
	20	$6.8 \times 10^{-1}$	125
	2	$6.7 \times 10^{-3}$	100
Base Case + 200 m <sup>2</sup> of SS at 300°C (Permeation Test)	200	$2.1 \times 10^1$	142
	20	$6.4 \times 10^0$	125
	2	$2.0 \times 10^0$	100
Base Case + 200 m <sup>2</sup> of SS at 300°C and BPT = 50 $\mu\text{Ci}/\text{m}^3$ (Back Permeation Test)	200	$2.1 \times 10^1$	142
	20	$6.0 \times 10^0$	125
	2	$1.6 \times 10^0$	100
Base Case + 20 m <sup>2</sup> at $\leq 0^\circ\text{C}$	20	$6.3 \times 10^1$	Tailing

<sup>a</sup>BPT = the average equivalent concentration of tritium in the high-temperature stainless steel (SS) that is contributing to back-permeation of tritium into the building. (See reference 8 for details of the calculations.)

During other experiments, similar to those in Fig. 3, one or the other, and in some cases both, of the processing steps (oxidation and water stripping) were temporarily by-passed. When both processing steps are by-passed at a point in time after tailing has set in, the tritium level in the enclosure usually tends to rise slowly with time, sometimes increasing overall by as much as 50 to 100%. Further, most of the residual tritium that is present after the tailing point has been reached can be removed from the enclosure simply by reinserting the molecular sieve bed alone. This result implies that, near the end of the simulated cleanup operation, a large fraction of the tritium present in the enclosure is in the oxidized form (HTO)—perhaps (as mentioned previously) the result of incomplete trapping of water by the molecular sieve bed.

**Computer Fitting of the Bench-Scale Data**

Following the experimental studies described in the preceding section, an attempt was made to derive reaction rate constants, HTO adsorption coefficients, and HTO desorption coefficients for the enclosure cleanup curves using TSOAK. By properly adjusting these three parameters it was possible to obtain exact fits to the experimental curves. The observed cleanup curves for two experimental cases are compared to the corresponding ideal cleanup curves in Fig. 4. Also shown in the figure are the data points for the TSOAK fit to the observed curve. (An asymptote of  $4 \text{ }\mu\text{Ci}/\text{m}^3$  was used in both the ideal and TSOAK calculations, and it is this asymptote which causes the curvature in the ideal curve.)

The experiment plotted on the left-hand side of Fig. 4 was performed by pulling clean air into the enclosure and exhausting the exit gas to a fume hood. This experiment, therefore, represents the condition of having a 100% efficient scrubbing system. The experiment plotted on the right hand side of Fig. 4 was performed under identical conditions (release size, flow rate, etc.) except that the enclosure atmosphere was circulated in a closed loop through the oxidizer bed and molecular sieve bed. The increased departure from ideal behavior in the latter case (compared to the once-through flushing case) is again attributable to a less than 100% HTO scrubbing efficiency of the molecular sieve bed. While on first through this appears to be the result of having an inadequately designed scrubbing system, it does not take much of a departure from 100% efficiency to create a sizeably increased apparent HTO production rate in the enclosure, i.e., less than 1% reduction in efficiency would be more than adequate to explain the differences in Fig. 4. Table 2 lists the TSOAK-derived fitting parameters for the two experiments in Fig. 4 and for two other interesting cases as well. Particularly worthy of note are the substantial increases in all three parameters when the fan and hot plate are both operating. The 400-fold increase in the required reaction rate parameter is considered to be well above the uncertainties associated with the computer model and with the experiment itself.

**Conclusions**

Although the results of the soaking studies described above are considered to be largely of semi-quantitative significance only, several conclusions can be drawn from the work carried out to date. These

Table 2  
SUMMARY OF CURVE MATCHING PARAMETERS OBTAINED FROM  
TSOAK FITS TO ENCLOSURE DETRITIATION EXPERIMENTS

Condition	Reaction Rate Parameter <sup>a</sup> (m <sup>3</sup> /μCi-min)	Adsorption Rate Parameter <sup>a</sup> (μCi/m <sup>2</sup> -min)	Release Rate Parameter <sup>a</sup> (%/min)
Typical Literature Data	10 <sup>-12</sup> -10 <sup>-14</sup>	--	--
Enclosure: Fan Off, Plate Cool	1 x 10 <sup>-5</sup>	3 x 10 <sup>-4</sup>	3
Enclosure: Fan On, Plate Cool	5 x 10 <sup>-5</sup>	2 x 10 <sup>-4</sup>	3
Enclosure: Fan On, Plate Hot <sup>b</sup>	4 x 10 <sup>-4</sup>	5 x 10 <sup>-5</sup>	12
Enclosure: Once-Through Flushing	1 x 10 <sup>-6</sup>	2 x 10 <sup>-4</sup>	3

<sup>a</sup>See reference 8 for a discussion of the derivation and implementation of these parameters.

<sup>b</sup>Plate heated to ~200°C.

may be stated summarily as follows: (1) When the relative humidity is greater than a few percent, the total amount of adsorbed tritiated water is greatly reduced due to surface swamping by H<sub>2</sub>O. (2) Exposed elevated-temperature metal surfaces (≥200°C) represent a major reaction and soaking pathway. All thermally hot metal surfaces should be insulated and preferably isolated from the building atmosphere. (3) Exposed low temperature surfaces (≥0°C) are also undesirable. Such surfaces (simulated by using sticking probabilities near unity) need not be large in area to condense large quantities of tritiated water. (4) For rooms in the range 10<sup>3</sup> to 10<sup>6</sup> m<sup>3</sup>, a 50 m<sup>3</sup>/s processor is desirable and an 0.5 m<sup>3</sup>/s processor is inadequate. With judicious selection of building materials, surface preparation procedures, and atmosphere operating parameters, a 5 m<sup>3</sup>/s system (near to state-of-the-art system) could be acceptable.

The principal value of predominantly empirical studies of the type described in this report is that they represent a medium with which results of actual detritiation experiments can be compared and correlated. Perhaps even more importantly they can (if properly used) provide a basis for the conception, design, and operation for meaningful large-scale air detritiation tests.

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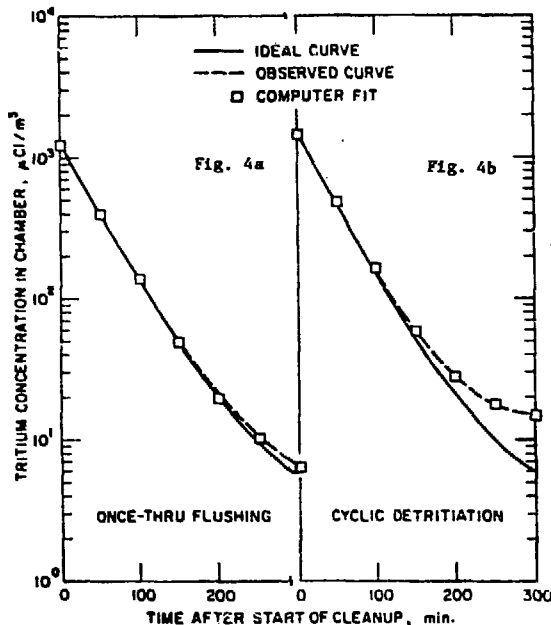


Fig. 4. Comparison of observed versus ideal and TSOAK calculated results for two air detritiation experiments (flow rate = 1 liter/min, enclosure volume = 50 liters)