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TRITIUM-CONTAINMENT SYSTEMS: A TRADEOFF STUDY

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ABSTRACT

We evaluated various design parameters that affect the performance of tritium-containment systems for fusion reactors. Our study included a review of such parameters as tritium forms, impurities, catalysts, adsorbents, getters, and "as low as reasonably achievable" principles. We organized these schemes, which can be considered for treating either air or inert atmospheres, so one could easily make orderly choices and tradeoffs for optimum performance. The relationships examined involved purification-system decontamination factors, flow rates, recycling and leakage, and environmental losses.

INTRODUCTION

Fusion-reactor facilities, which will handle large inventories of tritium, will have processing equipment that will inevitably suffer leakages and accidental losses. Containment systems must be provided to (a) minimize the tritium exposure of plant personnel and the general public and (b) recover the tritium for reuse or disposal.

The designer of tritium-containment systems must address an array of performance requirements. These range from government regulations concerning exposures and release rates to the effects of system leakage on the required design efficiency.

Some containment systems will be large and expensive, and thus poor designs could be costly if not dangerous.

This report especially concerns factors affecting overall system performance. We have organized these parameters so orderly choices and tradeoffs for optimum performance and cost can be easily made. Relationships involving purification-system decontamination factors, flow rates, recycling and leakage, and environmental losses are presented graphically.

RADIOLOGICAL STANDARDS AND DESIGN OBJECTIVES

Minimization of radiological exposures is the ultimate goal of containment systems. Radiological design guidelines provide the basis for establishing performance criteria for tritium-containment and -purification systems.

The Nuclear Regulatory Commission (NRC) also requires that the annual integrated population dose (man-rem) within 80 km (50 mi) of a source be

STANDARDS AND DESIGN OBJECTIVES

Fusion reactors can be expected to meet the radiation-exposure standards imposed on light water-cooled reactors [1-3] (Table 1). We are also expected to maintain "as low as reasonably achievable" (ALARA) exposures for workers and the general public.

TABLE 1. Radiation-exposure guidelines for light watercooled reactors.

Controlled area (mrem/yr)	Uncontrolled area (mrem/yr)
5000 (limit)[1]	–
–	25 (limit)[2]
–	5 (design objective)[3]

used to determine the environmental impact. NRC specifies a value of $1000/man-rem exposure to assess the cost/benefit of equipment to further reduce emissions. [3]

FENCELINE AND UNCONTROLLED-AREA EXPOSURES

The atmospheric dispersion of radionuclides following a release can be calculated by the procedures for noble-gas releases for light-water reactors. [4] We have arbitrarily chosen to use the "river site" location along with the normalized dispersion coefficients and population histograms given in Ref. 4.

Using the uncontrolled-area design objective of 5 mrem/yr for individuals at the site boundary, we calculated maximum release rates for 10- and 100-m stacks and the corresponding total population exposures (Table 2). Increasing stack height permits much larger maximum tritium-release rates, even though large reactor halls are unlikely to have stacks as low as this 10-m illustration.

Emissions of ~890 Ci/d from a 100-m stack will meet both the design objectives and the ALARA philosophy. Existing tritium facilities operate with emissions about a factor of 100 lower. [5,6] Estimates for fusion reactors are also a factor of 100 lower than calculated above. [7-9]

If we select an emission rate of 10 Ci/d from a 100-m stack, the fenceline exposure will be 0.06 mrem/yr, and the total population exposure will be 12 man-rem/yr. These lower values will fulfill the ALARA philosophy and can be achieved by present technology.

We also note that natural tritium production in the northern hemisphere is about ~ 70 Ci/d. [10] Hundreds of "10 Ci/d" plants could be operated before doubling the rate of atmospheric production.

TABLE 2. Maximum tritium-release rates for uncontrolled areas and total population exposures within 80-km (50-mi) radius for "river site."

Stack height (m)	Maximum release rate (Ci/d)	Total population-exposure rate/release rate (man-rem/yr)/(Ci/d)	Total population exposure (man-rem/yr)
10	6.4	6.0	38
100	890	1.2	1070

PURIFICATION SYSTEMS AND COMPONENTS

Purification systems can be designed to scavenge tritium either from air or from inert gasses. Several systems are operating successfully with air atmospheres, [5,11-14] and most reactor halls are expected to use air. Glove boxes often use inert atmospheres, [5,12,15] and at least one recent reactor design contemplates inert gas in a reactor hall. [16]

Catalytic oxidation—adsorption systems are used with air—tritium mixtures. These systems include the following features:

- Tritium may be scavenged as gas (HT), water (HTO), or organic vapors (termed CH_3T).
- Any gas with sufficient oxygen can be processed.
- Tritium is recovered at HTO.
- Contaminated air atmospheres will still be life-supporting for workers in an emergency.

Active metal (hydride) getter systems can be used with inert gas—tritium systems. These systems include the following features:

- Tritium is scavenged and recovered at HT, but HTO and CH_3T must be low.
- Inert atmospheres must be used.
- Systems can use packed or fluidized beds.
- Metal getters may be fire hazards if exposed to air.

CATALYSTS

Proven catalysts for oxidation of tritium include the precious metals platinum (Pt) and palladium (Pd) and mixtures of about 10 to 20 wt% of copper oxide in manganese dioxide called Hopcalite.* Both will oxidize HT and CH_3T, are more effective when heated, and are affected by poisons.

The most extensive kinetic data for oxidation of HT and CH_3T by precious metal catalysts is by

*Trademark of Mine Safety Appliances Company.

Bixel and Kershner.[17] Sherwood modified their reaction rate equation for pore diffusion: his estimates for catalyst efficiency as a function of temperature, residence time with the catalyst, and superficial velocity are shown for HT in Fig. 1.[18]
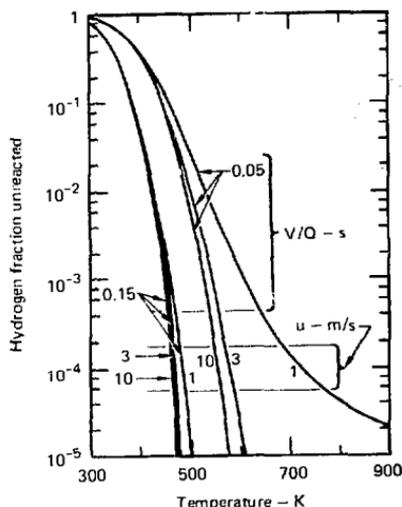

FIG. 1. Catalyst efficiency for two values of residence time, V/Q, and three values of superficial velocity, u. Calculated for Pt/A1$_2$O$_3$ catalyst bed with particle diam, d$_p$ = 3.18 mm and void fraction, E = 0.4 (Sherwood[18]).

Poisons affecting precious metal catalysts include mercury, halogens, silicones, carbon monoxide (CO), and sulfur compounds.[19-21] Hopcalite is affected by some halogens and CO.[19,22,23] In one installation, an activated charcoal filter scavenges poisons ahead of the catalyst.[12]

Hydrocarbons all require hot catalysts for oxidation, ranging from 150 to 415°C.[19,20] The lightest compounds [e.g., methane (CH$_4$) and ethane (C$_2$H$_6$)] require the highest temperatures.[19,24,25] Manufacturers's data for Hopcalite appear in Fig. 2.

Catalyst efficiencies apparently decrease at extremely low HT levels.[15,26] Some systems add

hydrogen (H$_2$) in an attempt to "swamp," or reduce, unoxidized HT leaving a catalyst bed.[13,27]

Circulation pumps may add energy to the gasses as heat of compression. This is used as preheat energy for better catalyst performance in several systems.[11,14,27]

Galloway noted the high cost of precious metal catalysts at about \$70 000/m^3 of catalyst.[7] He inferred a concern for costs by showing that cleanup time will vary inversely with the catalyst volume (Fig. 3).[28] The cost of Hopcalite is about \$7200/m^3 of catalyst.

ADSORBENTS

Common adsorbent materials include molecular sieves, activated alumina, and silica gel. The first two are capable of drying gasses to less than 0.1 Pa (~1 ppm) HTO; silica gel can dry gasses to about 1 Pa (~10 ppm) HTO. Adsorbent beds for tritium service are described by Sherwood[18] and Galloway.[9]

Adsorbent beds saturated with water (H$_2$O) can be used as isotopic exchange columns to scavenge HTO.[5,11,13,29] Tritium scavenged this way, however, will be diluted with large amounts of H$_2$O and will be difficult to recover for reuse.

Adsorbent beds can also be operated at cryogenic temperatures for direct adsorption of virtually all impurities, without requiring catalytic oxidation.[5,11]

PURIFICATION-SYSTEM PERFORMANCE

The most detailed performance results for a catalytic oxidation–adsorption system are reported by Gildea et al.,[14,30] and are reproduced in Fig. 4 for H$_2$ and CH$_4$. Sherwood reported one point for a comparable system with HT,[31] also shown in Fig. 4. The data show that the presence of any tritiated organics will require hot catalysts.

Two points are also shown in Fig. 4 for a catalytic oxidation−exchange column system using HT, by Flanagan, Rogers, and Wilkes.[11] We believe the general agreement with the data for adsorption systems is remarkably good.

The isotopic swamping technique noted earlier can also be used with adsorbent beds. One bed is
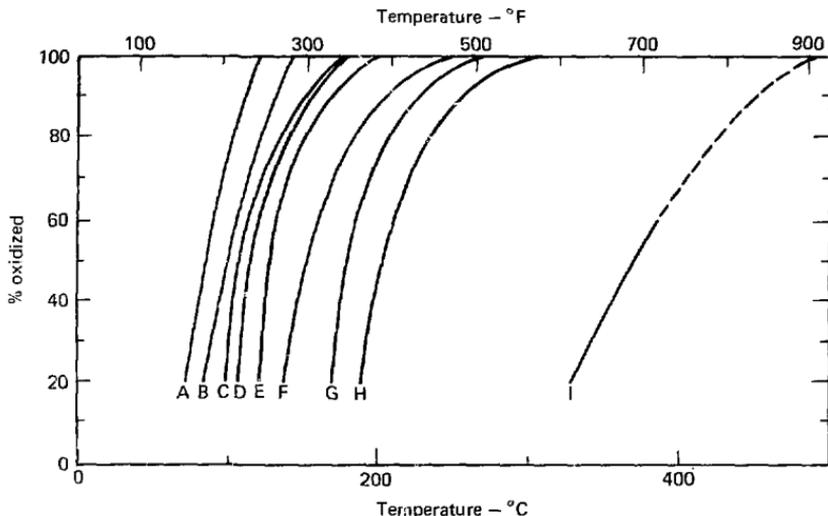
FIG. 2. Combustion of various contaminants over Hopcalite catalyst (Mine Safety Appliances part 26599) as a function of temperature (McDonough[25]). A = butene, C_4H_8; B = hexane, C_6H_{14}; C = acetylene, C_2H_2 and carbon monoxide, CO; D = propylene, C_3H_6; E = ethylene, C_2H_4; F = butane, C_4H_{10}; G =propane, C_3H_8 and hydrogen, H_2; H = ethane, C_2H_6 and benzene, C_6H_6; I = methane, CH_4.

used to adsorb HTO; then H_2O is added to the gas stream and a second bed brings down total moisture, resulting in lower HTO levels. [13,14,27,30,32]

GETTER MATERIALS

Active metal getters scavenge HT by forming stable metal hydrides. Candidates include uranium, titanium, zirconium, scandium, yttrium, thorium, and the rare earth series. Uranium and cerium were studied for use as packed beds. [33,34] Maienschein compared designs for packed and fluidized beds. [35]

An unsaturated organic material called DPPE* is also effective as a hydrogen getter and is reported stable in air. [36,37] The organic material is supported on calcium carbonate coated with a Pd catalyst.

CONTAINMENT-SYSTEM DESIGN REQUIREMENTS

MULTIPLE-CONTAINMENT DESIGN

The concept of multiple-containment barriers is generally accepted for tritium control in fusion reactors. [8,32,38-42] These are defined as primary, secondary, and tertiary confinement for high-level contamination, in the manner specified for certain other facilities. [43]
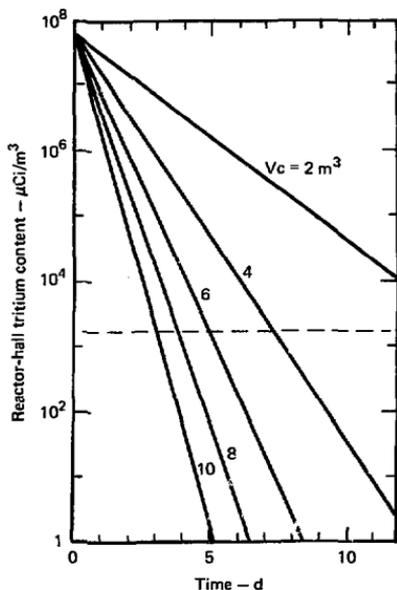
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*1,6-diphenoxy-2, 4-hexadiyne.

4

FIG. 3. Tritium reduction on "incredible accident," showing effects of catalyst volume, Vc, on cleanup time. Initial concentration of HTO (tritium scavenged as water) = 65 Ci/m^3; reactor-hall volume, V = 350 000 m^3; flow rate, Q = 58.8 m^3/s (10^5 cfm); temperature, T = 27°C (Galloway[28]).

FIG. 4. Purification-system decontamination factors for H_2 and CH_4.

Primary containment refers to the components and equipment in direct contact with tritium. Secondary containment refers to local containment enclosures surrounding pieces or sections of primary equipment. Tertiary containment includes the reactor hall and equipment rooms enclosing all tritium systems, including all containment purification systems.

The multiple-containment concept for a fusion-reactor plant can be illustrated by the diagram in Fig. 5, taken from Wittenberg, Wilkes, and Kershner. [8] Their study details containment concepts as well as explores tritium-source and -leakage terms.
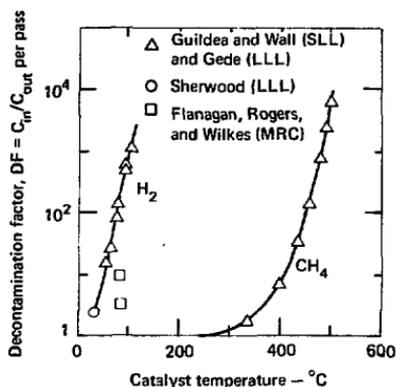
DESIGN PARAMETERS

Basic design features for catalytic oxi . tion-adsorption follow conventional chemical engineering design principles for oxidation by catalytic reactors and adsorption by packed beds.[44-48] We have already noted applications to tritium service by Sherwood[18] and by Galloway.[9] Aune, Cantelow, and Boltin[29] include design information for exchange columns. We will concern ourselves here more with special effects of system performance, recycling, leakage, and losses to the environment.

Most designs for reactor halls anticipate exponential dilution to low tritium levels. Sherwood derived a model showing the effects of tritium "soaking" into exposed surfaces and subsequent undesirable outgassing effects on this presumed exponential dilution.[49]

The relationships among the design parameters permit us to apply these functions as criteria for design. These include

● Reactor-hall volume, V (m^3).
● Purification-system flow rate, Q (m^3/s).
● Purification-system decontamination factor, DF = C_{in}/C_{out} per pass.
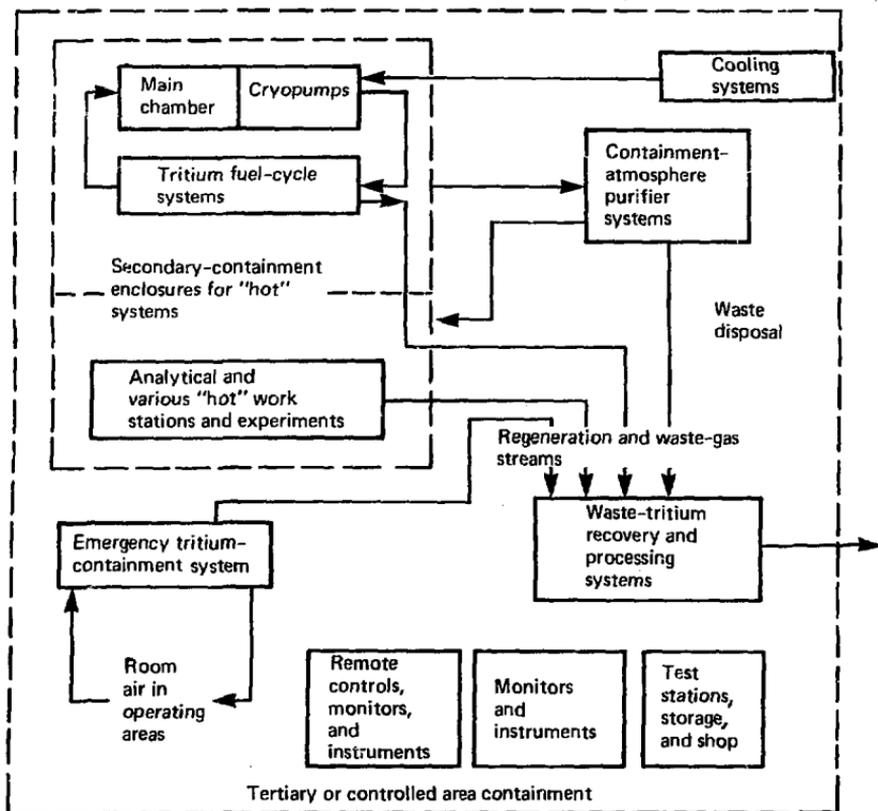
FIG. 5. Multiple-containment concept for tritium systems in a fusion reactor (*Wittenberg, Wilkes, and Kershner*[6]).

Make-up air

$C_{air} = 0$

$(1-R)Q$ Q

Reactor hall
V (m³)
M (Ci)
C (Ci/m³)

$C = C_{in}$
Q (m³/s)

Purification
system
DF = C_{in}/C_{out}

C_{out}
Q

**Leakage or
releases**

$(1-R)Q$

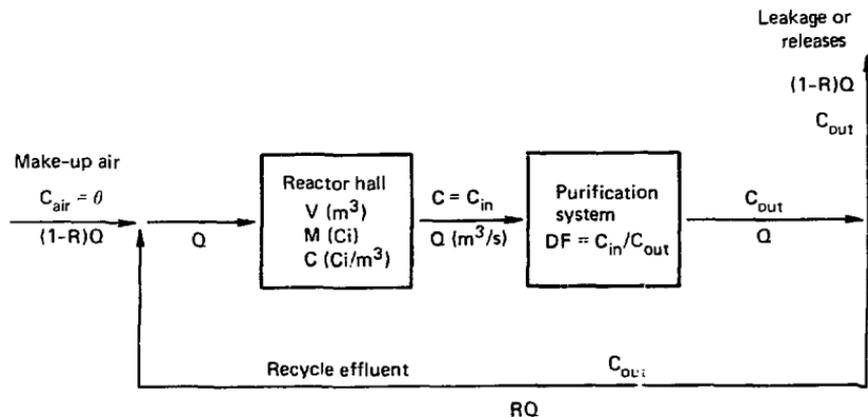C_{out}

Recycle effluent C_{out}

RQ

FIG. 6.   Purification-system flow pattern. Initial release = M_0 (Ci); initial concentration, $C_0 = M_0/V$ (Ci/m³).

- Fraction of the purification-system exhaust, R, that is recycled to the reactor hall (from 0 to 1.0).
- Time for recovery to a specified level, t (s).
- Reactor-hall concentration reduction factor, C/C_0, expressing the extent of cleanup for a reactor hall with any postulated release.
- Original tritium release, M_0(Ci); tritium remaining in the reactor hall, M (Ci); and integrated tritium release to the environment, M_L(Ci). Figure 6 shows the general flow patterns in any containment system (indicated as a reactor hall). For any initial release, M_0, we can show the concentration-reduction factor to be

$$\frac{C}{C_0} = e^{\left(\frac{R}{DF} - 1\right)\frac{Qt}{V}}, \qquad (1)$$

where $C_0 = M_0/V$. This can be plotted as a function of the dimensionless time, Qt/V, and is shown in Fig. 7 for several values of DF with R = 1.0. Note that the curve for DF = ∞ would also apply when R = 0.

The designer has several options:
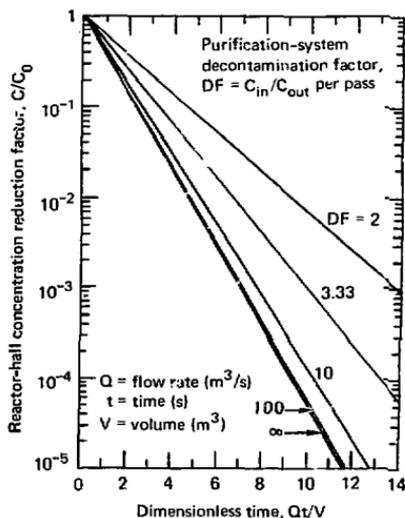- Values of V and DF can be traded to meet specified values of C/C_0 and t.

FIG. 7. Reactor-hall concentration reduction factor for several decontamination factors. Fraction recycled, R = 1.0; $C/C_0 = \exp\{[(R/DF) - 1] Qt/V\}$.

● A unit cost for a purification system with unit flow rate and a specified DF can be determined. The cost of the number of purifier units to meet specified values of C/C_0 and t can then be compared to the downtime cost of reaching the specified C/C_0. Increasing capital costs will provide lower (downtime) operating costs.

● Any two of the above parameters can be traded against each other, provided the other two parameters are specified.

The amount of tritium released to the environment, M_L, in the vented exhaust fraction, $1 - R$, is derived from the relations

$$\frac{dM_L}{dt} = (1 - R)QC_{out} \qquad (2)$$

for the rate of loss and

$$M = M_0 \, e^{\left(\frac{R}{DF} - 1\right)\frac{Qt}{V}} \qquad (3)$$

for the amount of tritium remaining in the reactor hall. Rearranging and integrating the rate of loss gives

$$M_L = \left[-\left(\frac{1 - R}{DF - R}\right) M_0 \, e^{\left(\frac{R}{DF} - 1\right)\frac{Qt}{V}} \right]_0^t \text{curies} \,. \qquad (4)$$

For large values of t this can be approximated by

$$M_L \cong \left(\frac{1 - R}{DF - R}\right) M_0 \text{ curies} \,. \qquad (5)$$

These results can be plotted as a fraction of the original release, M_L/M_0, vs the decontamination factor, shown for several values of recycle factor in Fig. 8.

Leakage of untreated gasses from a reactor hall or other tertiary containment can be controlled by operating at a slight negative pressure. The only releases are then in the fraction $(1 - R) Q$ as described above.

For any postulated release or accident, the designer can use the curves in Fig. 8 to trade system tightness, expressed as the fraction recycled, against system performance, expressed as the decontamination factor, to limit releases to the environment as required. Decontamination factors as low as 2 can be effective, given enough time and a tight system.

As an alternate method to minimize environmental releases, M_L, the effluent fraction $(1 - R)$ to be released can be treated by a smaller purifier having a high DF. The effluent fraction, R, to be recycled can be treated by less efficient purifiers, which are presumably less expensive to build or operate or both.
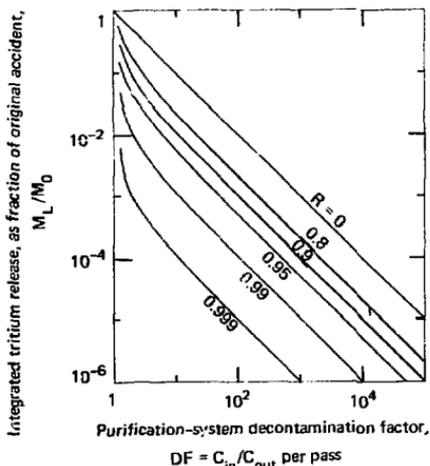

FIG. 8. Integrated tritium release to the environment for several recycle fractions. R = fraction of purification-system exhaust recycled to the reactor hall (0 to 1.0).

CONCLUSIONS

The radiological standards that will be applied to tritium releases from fusion reactors are more likely to be limited by ALARA principles than by off-site exposure design objectives. Existing technology can limit releases to less than 10 Ci/d, an emission rate that will generally result in off-site exposures well within the design objective.

We reviewed multiple-containment objectives and purification-system. components. Most tertiary containment systems are expected to have air atmospheres, requiring catalytic oxidation—adsorption systems. Tritium as HT can be effectively scavenged at or near room temperature. However, the likely presence of organically-bound tritium (CH_3T) or catalyst poisons (e.g.. CO) or both will require temperatures from ~350 to 500°C for precious metal and Hopcalite catalysts or else provision for replaceable filters to trap these materials.

Major parameters affecting system design are presented graphically. These figures will assist a designer in making orderly choices for optimum system performance and cost. We note that purification systems having only modest decontamination factors may be quite satisfactory, if all gasses are recycled and adequate cleanup time is allowed. However high decontamination factors are needed whenever the purified effluent is to be released to the environment.

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