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PERMEATION RATES FOR RTF METAL HYDRIDE VESSELS (U)

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May 21, 1992

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PERMEATION RATES FOR RTF METAL HYDRIDE VESSELS (U)

Contamination rates have been estimated for the RTF nitrogen heating and cooling system (NH&CS) due to tritium permeation through the walls of metal hydride vessels. Tritium contamination of the NH&CS will be seen shortly after start-up of the RTF with the majority of it coming from the TCAP units. Tritium contamination may elevate tritium concentrations in the NH&CS above 6×10^{-3} μCi/cc.

To reduce tritium activity in the NH&CS, a stripper or "getter" bed may need to be installed in the NH&CS. Increasing the purge rate of nitrogen from the NH&CS is shown to be an impractical method for reducing tritium activity due to the high purge rates required. Stripping of the NH&CS nitrogen in the glove box stripper system will give a temporary lowering of tritium activity in the NH&CS, but tritium activity will return to its previous level in approximately two weeks.

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SUMMARY

Contamination rates have been estimated for the RTF nitrogen heating and cooling system (NH&CS) due to tritium permeation through the walls of metal hydride vessels. Tritium contamination of the NH&CS will be seen shortly after start-up of the RTF with the majority of it coming from the TCAP units. Contamination rates of the NH&CS are estimated to exceed 400 Ci/year after three years of operation and will elevate tritium concentrations in the NH&CS above 6×10^{-3} μCi/cc.

To reduce tritium activity in the NH&CS, a stripper or "getter" bed may need to be installed in the NH&CS. Increasing the purge rate of nitrogen from the NH&CS is shown to be an impractical method for reducing tritium activity due to the high purge rates required. Stripping of the NH&CS nitrogen in the glove box stripper system will give a temporary lowering of tritium activity in the NH&CS, but tritium activity will return to its previous level in approximately two weeks.

INTRODUCTION

This report estimates the rate of tritium permeation from RTF metal hydride vessels into the RTF NH&CS. The impact these permeation rates have on the tritium activity in the NH&CS are discussed. These data were requested for determining the set point of the NH&CS Kanne chamber.

T. Mohler 5/27/92
Derivative Classifier

RTF process rooms will be evacuated when tritium activity exceeds 1×10^{-5} μCi/cc. It is desirable to maintain the tritium activity in the NH&CS below this level to minimize the risk to personnel in the event of a nitrogen leak into a process room. Stripping and purge control of the NH&CS nitrogen were proposed as methods for controlling tritium activity.

The calculations in this report include tritium permeation from the following metal hydride vessels: flow-thru beds, diffuser storage beds, recovery beds, Thermal Cycling Absorption Process (TCAP) feed beds, TCAP product beds, and TCAP. These calculations do not include the contributions from the primary and secondary hydride compressors since they have been eliminated from the scope of work for the RTF.

The transport of isotopes from the hydride vessels into the NH&CS is assumed to by permeation (diffusion) only. The model used to estimate permeation rates assumed isothermal conditions and constant physical properties at the calculation temperature. Pin-hole leaks in welds of the hydride vessel may significantly increase the rate of tritium transport into the NH&CS above what is estimated by permeation alone.

DISCUSSION

Permeation Calculations

Permeability of hydrogen isotopes through metal containers is a function of the concentration of the isotopes in the container and of the temperature of the container. A finite difference program developed at the Savannah River Site[1] for diffusion analysis of hydrogen systems was modified to examine permeation rates of tritium from RTF metal hydride vessels. This program calculated hydrogen isotope and helium concentration distributions through a structure wall as a function of the applied boundary conditions, the type of material used, the geometry of the vessel, the composition of the hydrogen isotopes, and the temperature of the vessel.

For the permeation calculations, the following conditions were used:

304L material of construction.

Cylindrical geometry.

The total pressure of hydrogen isotopes was constant inside the vessels, but the mole fractions of tritium/deuterium may differ. For the hydride beds, a total pressure of 2.5 atm was used since the mix tanks are typically limited to this value. For TCAP, the pressure varies and an average pressure of 3.0 atm of 100% tritium was used. These TCAP permeation data will be applied to 1/2 the total TCAP column area.

The partial pressures of hydrogen isotopes outside the vessel wall were taken to be zero.

3 inch schedule 10 and schedule 40 pipe were used for the hydride beds dimensions while 1-1/4" by 0.065" wall tubing was used for the TCAP dimensions.

A temperature of 165°C was used for the permeation calculations. This is the maximum hot nitrogen gas temperature expected in the annulus of the metal hydride vessels.

Tritium permeation rates for different pipe schedules and tritium/deuterium mixtures are shown in Figure 1. These results were calculated for the hydride vessels heated to their maximum temperature 24 hours per day. To calculate the permeation rate for heating at less than 24 hours per day, multiply the time scale of Figure 1 by (24/X), where X is the number of hours per day the bed is heated.

For an estimate of the permeation rates to be expected in the RTF, it was assumed that the hydride beds would be heated for a total of 3 hours per day and TCAP would be heated for 12 hours per day. This assumption is reasonable based on the expected operation of the RTF process and will be used throughout the remainder of this report.

The re-scaled permeation rates from Figure 1 are shown in Figure 2 for up to 20 years of operation. These data shown that permeation rates for schedule 40 pipe are significantly lower than permeation rates for schedule 10 pipe. Figure 2 also shows that tritium permeation from TCAP will be seen shortly after start-up of the RTF.

Permeation Surface Areas

The permeation surface areas for the beds were based on the outside diameter of 3 inch pipe while TCAP used the outside diameter of 1-1/4" tubing. For the beds, estimates included the surface area of the pipe caps assuming flat pipe caps. The

FIGURE 1. TRITIUM PERMEATION RATES FOR RTF HYDRIDE VESSELS
VESSELS HEATED 24 HOURS/DAY

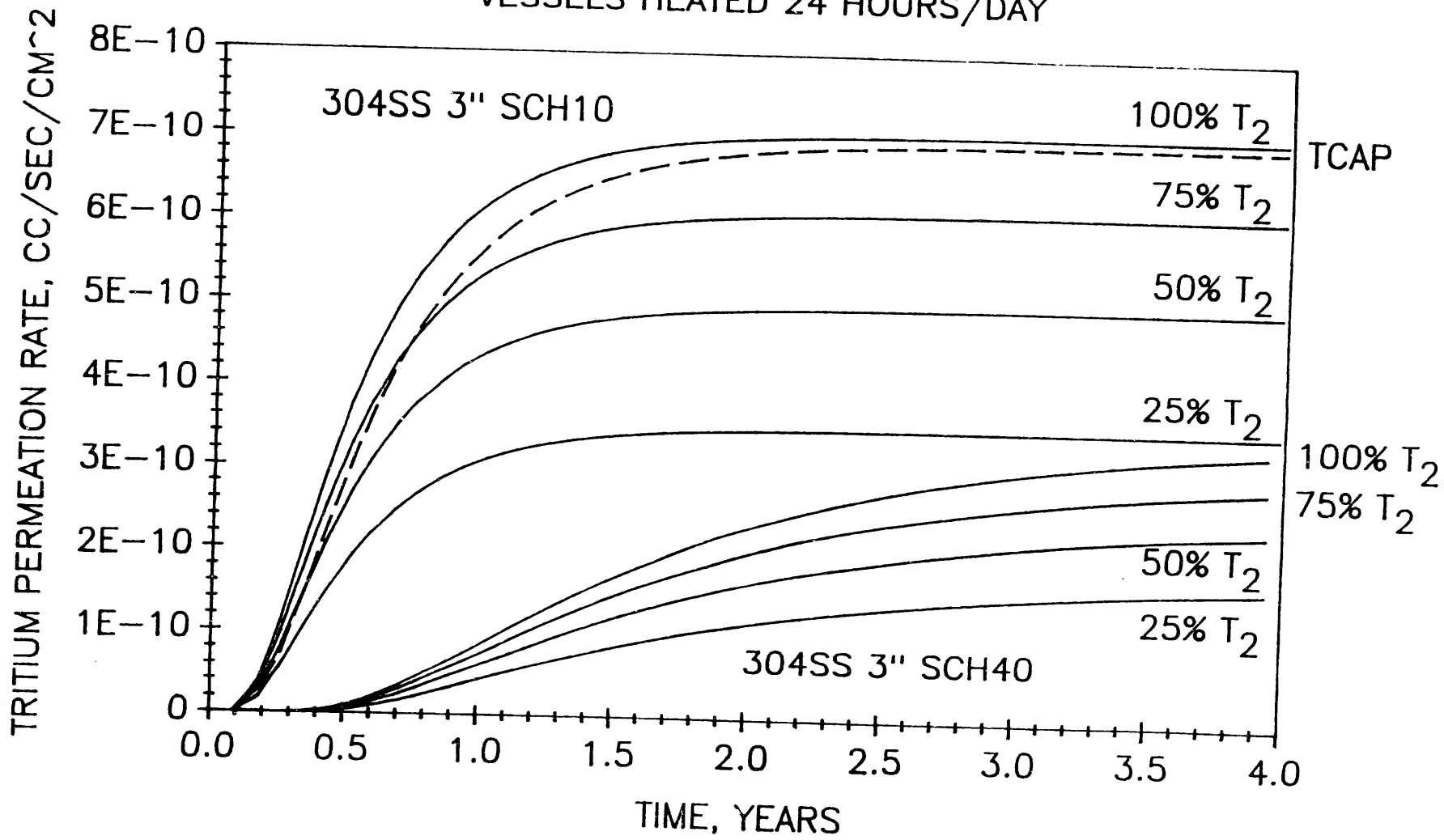
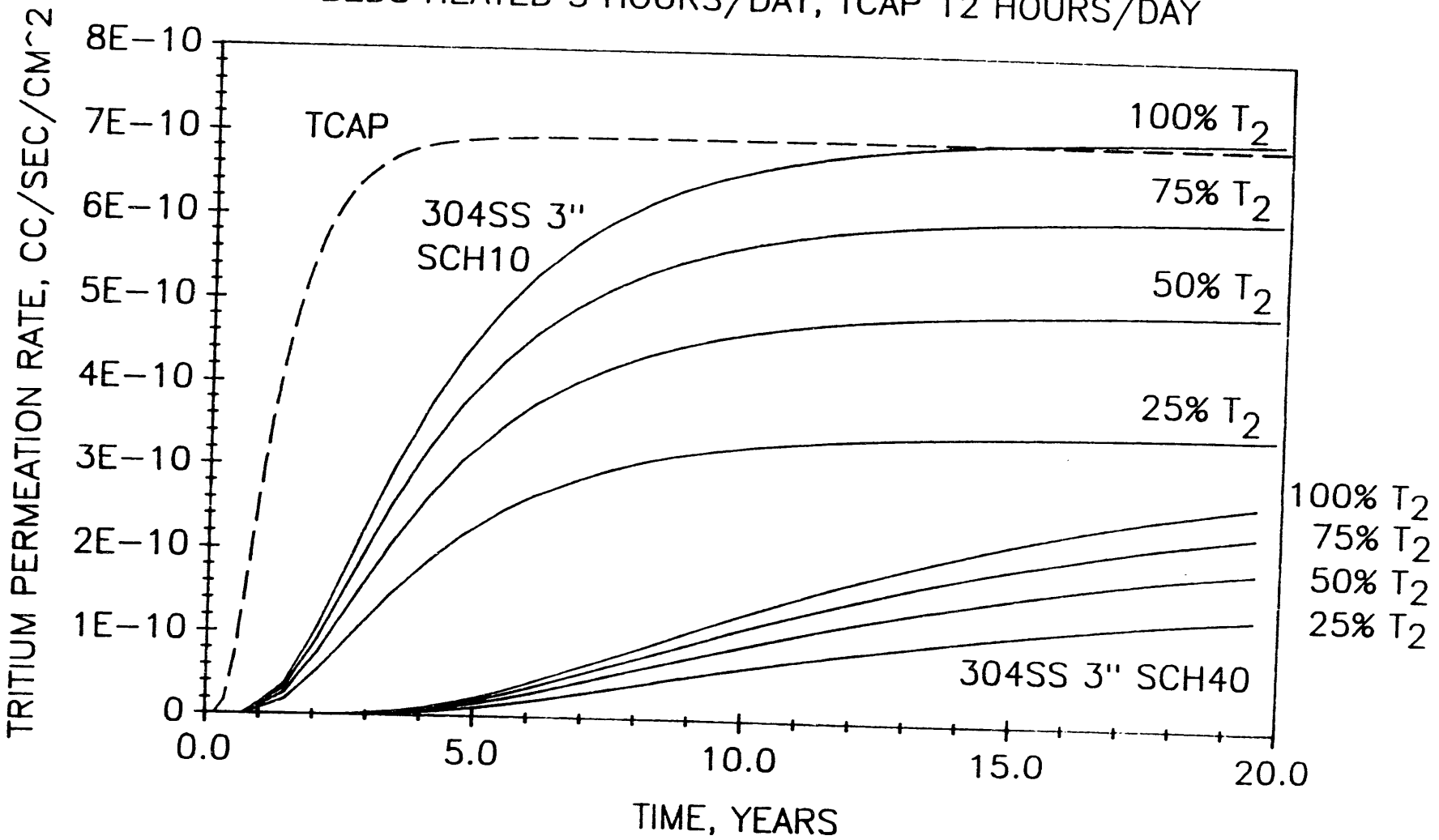


FIGURE 2. TRITIUM PERMEATION RATES FOR RTF HYDRIDE VESSELS
BEDS HEATED 3 HOURS/DAY, TCAP 12 HOURS/DAY



equation used for the beds was

$$Area \approx \pi DL + (2) \times \frac{\pi D^2}{4}$$

where D is the outside diameter of the pipe and L is the approximate length of the vessel.

Storage Bed Areas

Storage beds used in the RTF include the diffuser storage beds, recovery beds, TCAP feed beds, and TCAP product beds (TCAP raffinate beds will not see significant amounts of tritium).

3 inch Sch10 pipe, L ≈ 36 inches long

Area ≈ 2678 cm^2/bed with a total of 16 beds

Flow-Thru Bed Areas

3 inch Sch10 pipe, L ≈ 60 inches long

Area ≈ 4380 cm^2/bed with a total of 6 beds

TCAP Areas

The feed to the TCAP is assumed to be 50% tritium so only 1/2 of the TCAP column will be exposed to tritium and the calculated area will be divided by 2.

1-1/4 inch by 0.065 inch wall tubing, L ≈ 40 feet long

Area ≈ 12,161 cm^2/TCAP with a total of 2 TCAP units

Contamination Calculations

The contamination rate is defined as the rate at which tritium is transported into the NH&CS. This is equal to the permeation rate for a vessel times its permeation surface area. To get the total contamination rate for the NH&CS, the contamination rate is summed over all the hydride vessels.

To compensate for the fact that the hydride vessels are not heated at all times, the contamination rate is calculated using

$$C_{rate} = P \times Area \times 3600 \times \frac{X hours}{day} \times 365.25 \times 2.57 \frac{Ci}{cm^3}$$

where C_{rate} is the contamination rate in Ci per year per vessel, P is the permeation rate in SCC tritium per cm^2 surface area per sec, X is the number of hours per day the vessel is at its maximum temperature, taken to be 3 for the hydride beds and 12 for TCAP, and the other numbers are for unit conversions.

For RTF operations, it will be assumed that the gas composition present in the hydride vessels preceding TCAP are 50% tritium with the balance being deuterium. With 50% tritium feed to TCAP, it is assumed that 1/2 the TCAP column will be exposed to 100% tritium during the heating cycle. For the TCAP product beds, the gas composition will be assumed to be 100% tritium.

Figure 3 shows the contamination rates for different tritium compositions and different hydride beds. Additional data are included in this figure to show how different schedule pipe affects the contamination rate for a particular type of bed. Figure 4 shows the contamination rate for TCAP which is larger than for any of the individual beds.

Contributions to the contamination rates from all the flow-thru beds at 50% tritium, the storage beds at 50% tritium, the storage beds at 100% tritium, and the TCAP units are shown in Figure 5. Figure 5 also shows the total contamination rate with the TCAP units contributing the major portion of the total contamination rate.

Uncertainties in the permeation calculations will arise from the assumption of constant tritium partial pressure, constant temperature and the amount of time these conditions are maintained. After break-through, the permeation rate is approximately proportional to the partial pressure of tritium contained inside the hydride vessel. Reduction of the tritium partial pressure by a factor of two inside a vessel will decrease the contamination rate from the vessel by approximately 1/2.

An Arrhenius type expression is used to estimate the diffusivity of hydrogen isotopes through stainless steel. By reducing the temperature used in the permeation calculations from 165°C to 150°C, the contamination rate for the vessels would be reduced by almost a factor of 2. Reducing the time the vessels are heated to these temperatures will also reduce the calculated contamination rate.

FIGURE 3. TRITIUM CONTAMINATION RATES FOR RTF HYDRIDE VESSELS
BEDS HEATED 3 HOURS/DAY

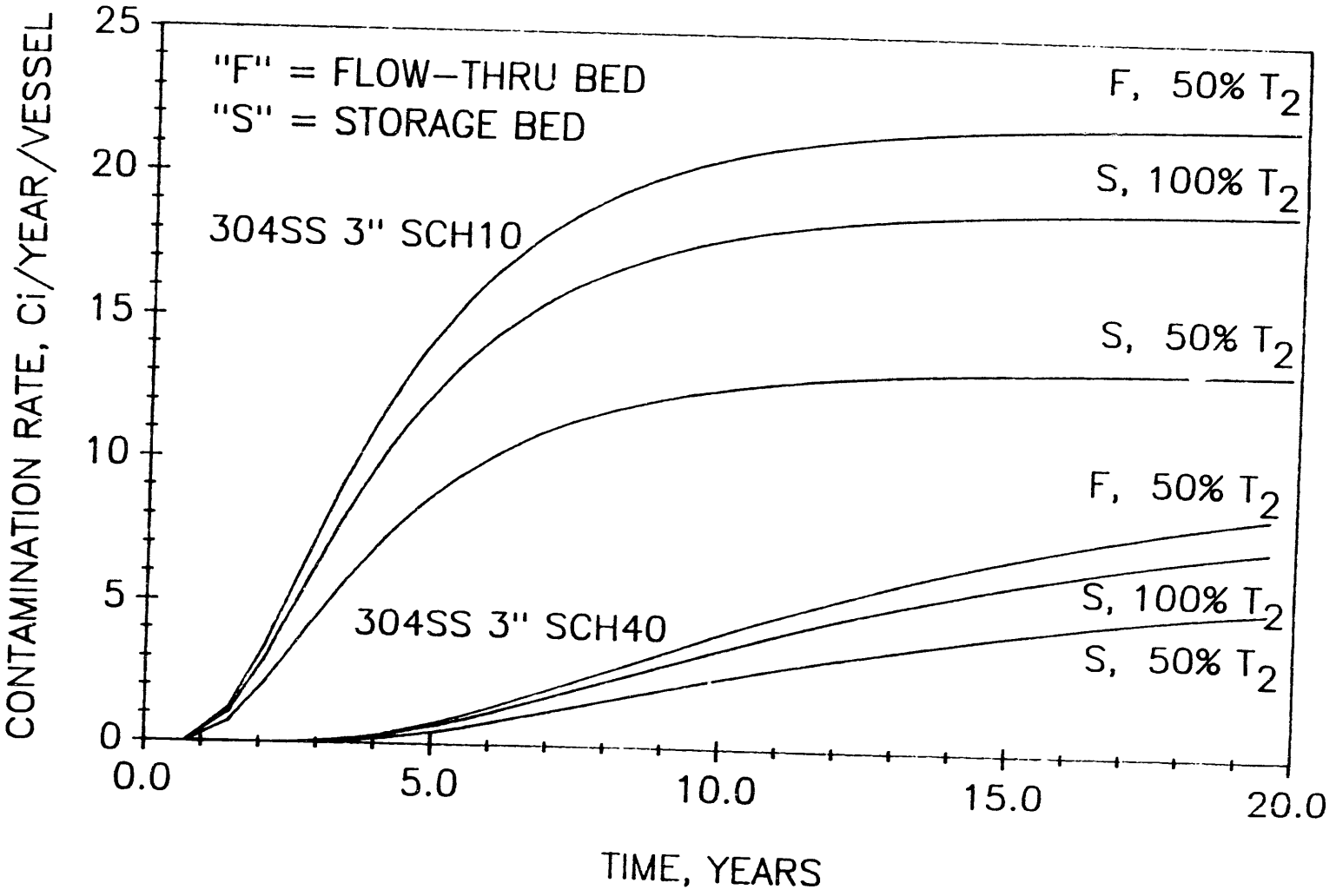


FIGURE 4. TRITIUM CONTAMINATION RATES FOR RTF HYDRIDE VESSELS

TCAP HEATED 12 HOURS/DAY

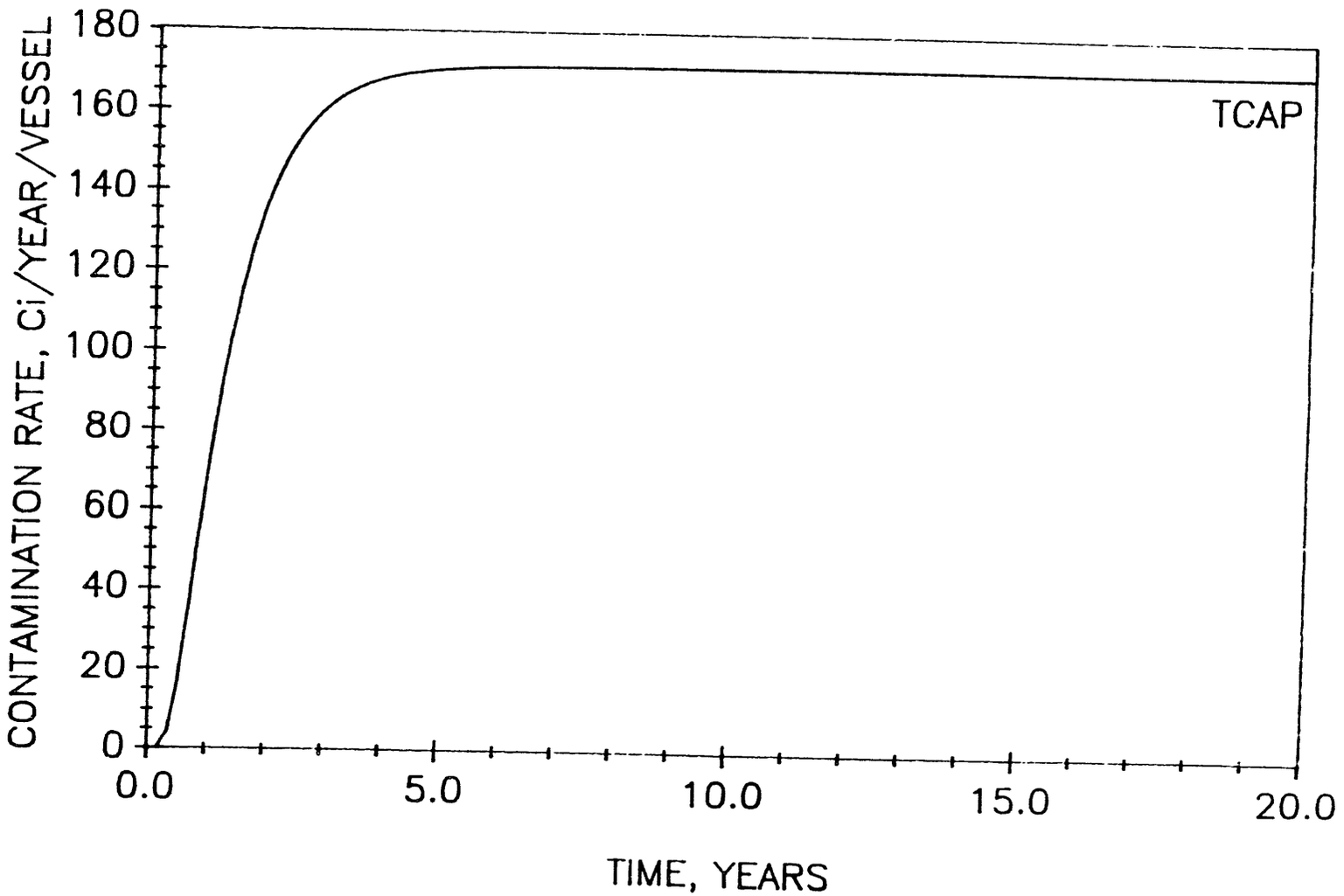
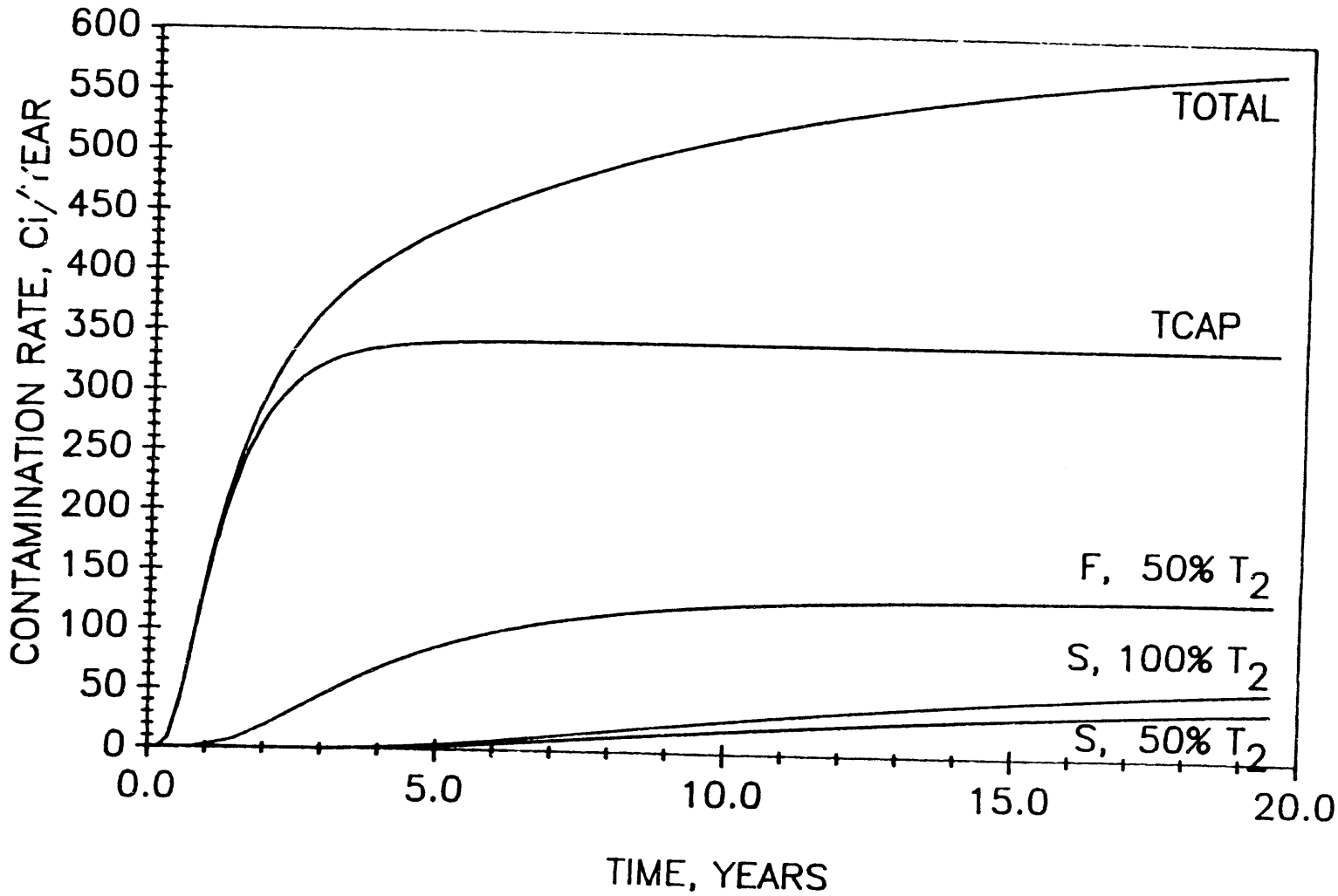


FIGURE 5. TRITIUM CONTAMINATION RATES FOR RTF HYDRIDE VESSELS
BEDS HEATED 3 HOURS/DAY, TCAP 12 HOURS/DAY



NH&CS Tritium Concentration

The concentration of tritium in the NH&CS can be estimated by treating the NH&CS as a continuous stirred tank reactor (CSTR). Nitrogen inputs to the CSTR includes nitrogen flow from the compressor seal over-pressure system and from nitrogen supplied through the nitrogen make-up system. For steady-state operation, the purge rate from the CSTR must be the same as the inlet flow and is controlled by the flow rate through the purge stripper. The tritium input term for the CSTR is assumed to come only from tritium permeation.

For some initial tritium concentration in the nitrogen system, C(0), the transient response for the CSTR model is given by

$$C(t) = \frac{Q}{F}\left(1 - e^{-\frac{t}{T}}\right) + C(0)\, e^{-\frac{t}{T}}$$

where Q is the tritium contamination rate into the system, F is the steady-state nitrogen flow (purge) rate through the system, t is time, and T is time constant for the system and is equal to the volume of the NH&CS, taken to be 3600 SCF, divided by F.

The steady-state tritium concentration in the NH&CS is given by the ratio Q/F. For a purge rate of 1 SCFM, the time constant T is equal to 2.5 days and any transient in the above equation will be negligible after two weeks.

The contamination rates from Figure 5 were used to calculate steady-state tritium concentrations in the NH&CS as a function of time and are shown in Figure 6 for different purge rates. After one year of operation, the nitrogen purge rate can be used to control the tritium activity to between 6×10^{-3} μCi/cc and 6×10^{-2} μCi/cc. These activities are of the same magnitude as the activities expected in the nitrogen glove atmosphere.

To show if increasing the nitrogen purge rate through the NH&CS can be used to reduce tritium activity, activity in the NH&CS as a function of nitrogen purge rate is shown in Figure 7. Figure 7 clearly shows that increasing the nitrogen purge rate through the system is not a practical way to reduce tritium activity in the NH&CS since the purge/make-up rate is limited to approximately 5 SCFM.

To clean-up the tritium in the NH&CS in the stripper system, twelve hours will be required. This is the time required to empty the NH&CS nitrogen inventory through the stripper system (limited by the purge stripper at 5 SCFM). As stated before, even after re-filling the NH&CS with fresh nitrogen, steady-state

FIGURE 6. NITROGEN SYSTEM TRITIUM ACTIVITY

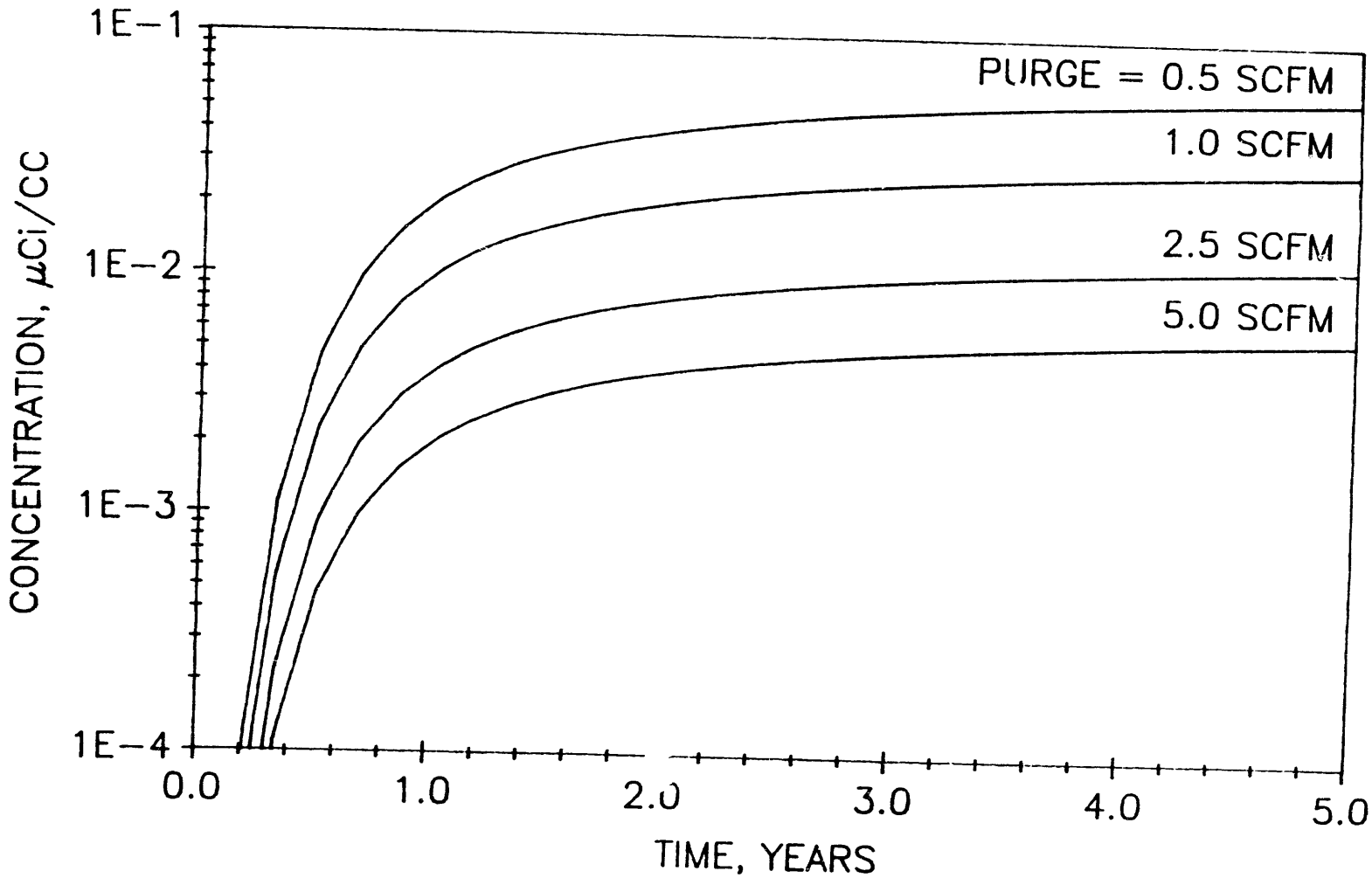
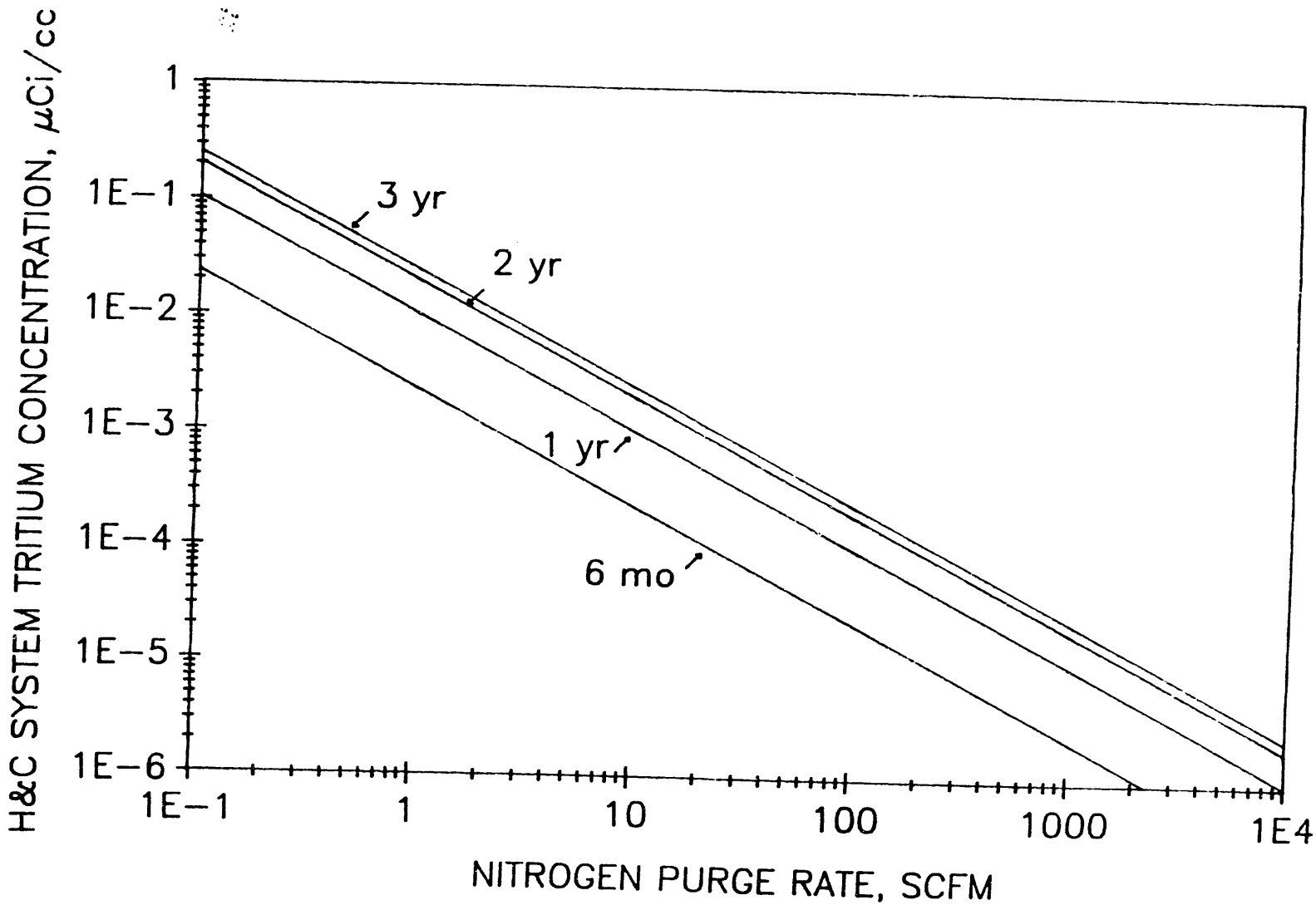


FIGURE 7. H&C NITROGEN SYSTEM TRITIUM ACTIVITY



tritium concentrations will return to previous levels in approximately two weeks.

CONCLUSIONS

Tritium permeation through RTF metal hydride vessels will become significant shortly after start-up of the RTF. TCAP is the major contributor to the total tritium contamination rate experienced by the NH&CS. Increasing the nitrogen purge rate through the NH&CS is not a practical method of reducing tritium activity levels in the NH&CS. Stripping tritium from the NH&CS nitrogen using the strippers present in the RTF will take at least 1/2 of a day and will only temporarily reduce tritium activity in the NH&CS.

To reduce tritium activity levels in the NH&CS, some type of tritium stripper or "getter" system may need to be installed into the NH&CS. Several potential tritium getter materials have been identified and are to be tested in the SRTC. Additional steps should also be taken to reduce permeation rates of tritium through the walls of TCAP via alternate TCAP design.

REFERENCES

1. Kain, K. E. *Finite-Difference Program for Hydrogen Diffusion*. USDOE Report DP-1738, Savannah River Site, Aiken, SC 29808 (1987).

END

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