

EGG-M-90213
CONF-90/007-36

TRITIUM PERMEATION LOSSES IN HYLIFE-II HEAT EXCHANGER TUBES^a

EGG-M--90213

Glen R. Longhurst and Thomas J. Dolan
Idaho National Engineering Laboratory
P.O. Box 1625, Idaho Falls, ID 83415-3523
(208) 526-9950 (208) 526-1649

DE91 006136

ABSTRACT

Tritium permeation through the intermediate heat exchanger of the HYLIFE-II inertial fusion design concept is evaluated for routine operating conditions. The permeation process is modelled using the Lewis analogy combined with surface recombination. It is demonstrated that at very low driving potentials, permeation becomes proportional to the first power of the driving potential. The model predicts that under anticipated conditions the primary cooling loop will pass about 6% of the tritium entering it to the intermediate coolant. Possible approaches to reducing tritium permeation are explored. Permeation is limited by turbulent diffusion transport through the molten salt. Hence, surface barriers with impedance factors typical of present technology can do very little to reduce permeation. Low Flibe viscosity is desirable. An efficient tritium removal system operating on the Flibe before it gets to the intermediate heat exchanger is required. Needs for further research are highlighted.

INTRODUCTION

HYLIFE-II is a design study to examine the feasibility of replacing the molten Li coolant in the HYLIFE inertial confinement reactor design concept with a mixture of LiF and BeF₂ known generically as Flibe. Part of that study deals with the power system and particularly the requirements for tritium management to prevent unacceptable losses of tritium to the environment through it. The objective for tritium releases is 40 Ci/day to achieve acceptably low radiation doses at the site boundary.

Tritium is bred in the Flibe by neutron interactions with the Li. The design of the effectively black to neutrons produced in the blast. Hence, with some neutron multiplication, the tritium production in the Flibe is a little reactor is such that the Flibe curtain is

^aWork supported by U.S. Department of Energy, Office of Energy Research under DOE Contract DE-AC07-76ID01570.

larger than the tritium consumption, the difference being mainly due to (n,2n) reactions in the Be. For a fusion thermal power of 2800 MW_{th}, the anticipated tritium load in the Flibe is about 4.8 MCi/day.

A major pathway for the escape of this tritium to the environment is through the heat exchangers. The very design characteristics that make an effective heat exchanger (narrow tubes, thin walls, high temperature, and turbulent flow) also foster the ready loss of tritium through the heat exchanger tubes. Depending on the circumstances, the rate of that loss can be influenced by many factors including temperature, material properties, and flow conditions. Diffusive transport through membranes has been extensively studied, but in the case of turbulent flow of molten salt laden with tritium, there has been little work done to evaluate the driving potential to use in diffusion calculations. It is apparent that because of the flow turbulence, there will be a boundary layer in which the dissolved tritium concentration is lower than in the free stream. It has not been clear, though, how much lower it will be. An additional complication is the tendency of the permeation flux to become proportional to the driving potential to the first power rather than to its half power as is usually the case in diffusion problems. The details of how and where this transition takes place need clarification.

The objective of this study is to develop a methodology for analyzing tritium losses in this type of heat exchanger and then to use the model developed to evaluate potential design features in terms of their effect on tritium loss rates.

ANALYTICAL MODEL

The driving potential for permeation of tritium through a membrane can be expressed in terms of a gaseous pressure at the membrane surface, P₁. The net rate of movement of tritium molecules into a metal surface is the difference between the rates of dissociation and recombination at that surface. Dissociation is modelled as the product of a dissociation coefficient, K_d, and the

Received by OSTI

JAN 15 1991

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

pe

driving pressure, P_1 . In the absence of a species chemically receptive to tritium at the surface, the recombination rate is $K_r c^2$ where c is the dissolved tritium concentration at or near the surface of the metal. The equality of the dissociation and recombination terms under no-flow conditions leads to the conclusion that $K_d = S^2 K_r$, where S is the solubility of tritium in the metal. When the flow of tritium through the membrane is fully diffusion limited, we assume that at the upstream side the concentration, $c_1 = S \sqrt{P_1}$ as given by Sieverts' law while at the downstream side c_2 vanishes. The presence of a tritium partial pressure on the downstream side of the membrane results in a back flow that can be superimposed on the solution for forward diffusion. The resulting equation for the net permeation flux density, J , may be written as

$$J = \frac{\phi S D}{b} \left(\sqrt{P_1} - \sqrt{P_2} \right) \quad (1)$$

Here D is the diffusivity of tritium in the metal, and ϕ represents a degradation factor due to the influence of surface recombination processes (diffusion no longer governs completely). We also define a permeation parameter, W , similar to those of Waegbroeck et al.,¹ Kerst and Swansiger,² Doyle,³ and others that is essentially the ratio of diffusive impedance to surface-limited impedance to permeation:

$$W = \frac{S b K_{r1}}{D} \left(\sqrt{P_1} - \sqrt{P_2} \right) \quad (2)$$

The parameter, ϕ , may be thought of as a dimensionless permeation flux. By making use of surface dissociation and recombination processes together with Fick's law of diffusion, the relationship between ϕ and W can be shown to be given by the transcendental relation

$$\phi = \sqrt{\beta - \frac{\phi}{2W}} - \sqrt{\frac{P_2}{P_1} \beta + \alpha \frac{\phi}{2W}} \quad (3)$$

where

$$\beta = \frac{1}{\left[1 - \sqrt{\frac{P_2}{P_1}} \right]^2} \quad (4)$$

and α is the ratio of upstream to downstream recombination coefficients. This functional relationship is shown graphically for $\alpha = 1$ in Figure 1. It will be seen there that when W is large (diffusion limit), ϕ approaches unity, and

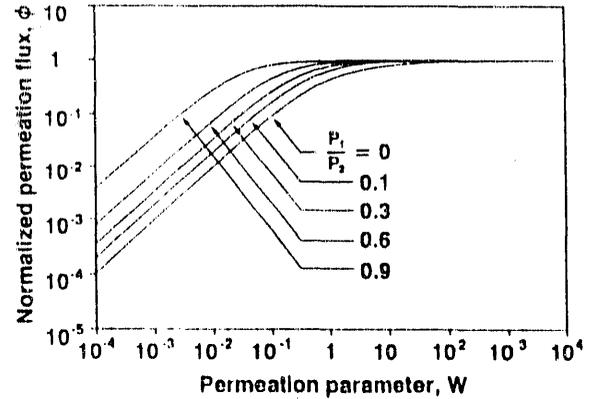


Figure 1. Relation of normalized permeation flux to permeation parameter, W , for a range of pressure ratios.

permeation is proportional to the half power of driving pressure. In the limit of small W (recombination limit), ϕ becomes proportional to W which means the permeation flux, J , is proportional to driving pressure to the first power (see Eqs. (1) and (2)). Increasing the downstream pressure reduces the value of W at which the transition to $\phi = 1$ occurs. Increasing α moves the curve set sharply to the right while reducing it moves it left only slightly.

The rate of tritium loss through heat exchanger tubes will be strongly dependent on the chemical potential or the effective pressure, P_1 , at the tube surface. To determine what P_1 should be, we apply the Lewis analogy that implies dissolved tritium concentration profiles with the same shape as momentum or temperature profiles, assuming turbulent flow.⁴ Specifically, a reference velocity, v^* , is defined by

$$v^* \equiv 0.1989 u Re^{-1/8} \quad (5)$$

where u is the average stream velocity, and Re is the conventional Reynolds number for flow in a tube. A dimensionless concentration, c^* , of dissolved tritium in Flibe is then defined by

$$c^*(r) \equiv \frac{v^*}{J} [\hat{c}(r) - \hat{c}(R)] \quad (6)$$

where $\hat{c}(r)$ is the corresponding dimensional value. Next a dimensionless distance, r' , from the tube wall is defined that is a form of Reynolds number based on the reference velocity:

$$r'(r) \equiv \frac{v^* \rho}{\mu} (R - r) \quad (7)$$

where R is the tube radius, ρ is the Flibe mass density, and μ is its viscosity. For $r^* < 26$ the dimensionless concentration profile is given by

$$c^*(r^*) = \int_0^{r^*} \frac{ds}{\frac{1}{Sc} + nu^*s[1 - \exp(-nu^*s)]} \quad (8)$$

where Sc is the Schmidt number

$$Sc = \frac{\mu}{\rho D_f} \quad (9)$$

while μ is the Flibe viscosity⁵ and D_f is the diffusivity of tritium in Flibe.⁶ The accepted value of n in Eq.(8) is 0.124 and

$$\begin{aligned} u^* &= s, & s \leq 5 \\ u^* &= 4.731 \ln(s) - 2.664, & s > 5 \end{aligned} \quad (10)$$

For $r^* > 26$

$$c^*(r^*) = \frac{1}{0.36} \ln \left(\frac{r^*}{26} \right) + c^*(26) \quad (11)$$

Denoting by $\langle c^* \rangle$ and $\langle \hat{c} \rangle$ the radially averaged values of c^* and \hat{c} , and applying continuity and Henry's law, Eq.(1) may be combined with Eq.(6) to give

$$\frac{d\langle \hat{c} \rangle}{dz} = - \frac{v^*}{\langle c^* \rangle Ru} (\langle \hat{c} \rangle - P_1 \hat{S}) \quad (12)$$

where R is the tube radius and \hat{S} is the solubility of tritium in the Flibe. Recognizing that $P_1 \hat{S}$ is really just the concentration of tritium in the Flibe at the wall and that in many instances this is much less than $\langle \hat{c} \rangle$, Eq.(12) may be reduced to

$$\langle \hat{c} \rangle = \hat{c}_0 \exp \left[- \int_0^z \frac{v^*}{\langle c^* \rangle Ru} dz \right] \quad (13)$$

where \hat{c}_0 is the concentration at the tube inlet. Solution of either Eq.(12) or Eq.(13) is by Runge-Kutta integration. We ignore boundary layer development effects at the tube entrance. Then, beginning with a known initial value of P_1 with P_2 assumed fixed and known material properties, we are able to estimate a value for W via Eq.(2). That in turn leads to ϕ through Eq.(3). Recasting Eq.(6) as

$$\langle c^* \rangle = \frac{bv^*}{\phi SD (\sqrt{P_1} - \sqrt{P_2})} (\langle \hat{c} \rangle - P_1 \hat{S}) \quad (14)$$

and with current values for P_1 , $\langle \hat{c} \rangle$ and ϕ , $\langle c^* \rangle$ may be found and used with Eq.(12) to obtain the derivative for the next integration step. We used the Baskes⁸ recombination model in our solution for W and assumed that the temperature of the flibe near the wall was at the mean of the wall and bulk flibe temperatures.

APPLICATION

The general layout of the HYLIFE-II power system is shown in Figure 2. Tritium is bred in the flibe of the reactor vessel at the rate of about 4.90 MCi/day. Nominally 95% of the flibe is recirculated through the vessel to provide shielding for the walls while 5% passes through a vacuum disengager before going through the intermediate heat exchanger (IHX). Part of that flow is passed through a Be contactor to control flibe chemistry. The vacuum disengager system prior to flibe entry into the IHX is for the removal of tritium. Several IHX designs have been considered, but the one chosen for evaluation here has 7,400 tubes, each 10 m long and 2 cm in outside diameter with an assumed thickness of 1 mm. The flibe flow rate to the IHX is 2 m³/s for an average tube velocity of about 1.1 m/s. Inlet temperature is 943 K, outlet temperature is 843 K, and on the average the wall is assumed to be about 75 K cooler than the bulk flibe temperature. We assumed that the gas exchange column in the intermediate NaBF₄ loop removes enough tritium that the downstream pressure, P_2 for the IHX tubes is negligible (10^{-8} Pa was used). Baseline tube material is type 316 stainless steel.

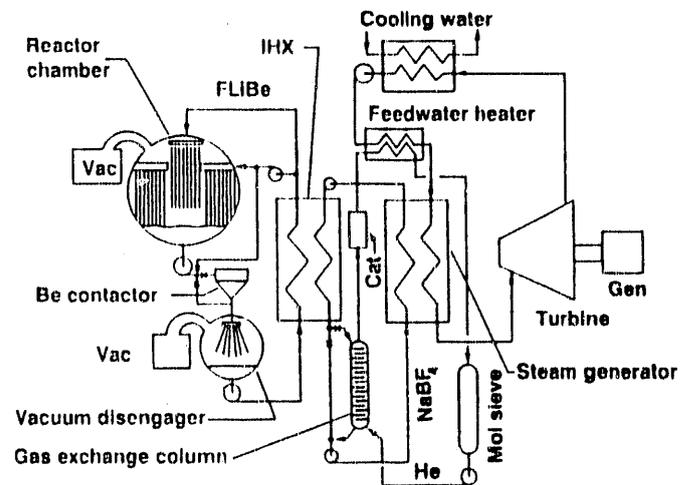


Figure 2. Schematic showing the major elements of the HYLIFE-II power and tritium control systems.

EVALUATION

Calculations using a computerized algorithm and simplified hand calculations both gave the permeation fraction for tritium passing through the IHX of about 6%. That fraction was found to be virtually independent of the average tritium partial pressure present. To establish sensitivities to other design parameters, additional runs were made, the results of which are listed in Table 1.

Table 1. Results of calculations to examine sensitivity of tritium permeation to IHX design options.

Feature	Loss (%)	Permeation Param., W	Edge/Bulk Conc. Ratio	Outlet P_{12} (Pa)
Baseline	6.02	1.77E-02	2.86E-08	7.1E-07
$K_r \times 10^{-5}$	6.02	1.27E-04	1.14E-04	2.9E-03
$u \times 2$	5.52	2.49E-02	5.25E-08	1.3E-06
$b \times 5$	6.02	9.43E-02	3.20E-08	7.9E-07
$R \times 5$	1.00	1.62E-02	2.33E-08	6.1E-07
$H \times 2$	11.69	1.71E-02	2.86E-08	6.7E-07
$T + 100 \text{ K}$	11.72	2.09E-02	5.13E-08	1.1E-06
Vanadium	5.94	3.04E-06	4.15E-10	1.0E-08
Tungsten	6.02	1.92E-03	6.90E-09	1.7E-09
Composite	5.94	2.56E-07	4.15E-10	1.0E-08

For each of the calculations tabulated here, the tritium partial pressure at the inlet to the tube was taken as 13.3 Pa or 1 torr. That is the nominal design point for the vacuum disengager pumping system. However, the choice of inlet partial pressure did not impact loss fraction. The permeation parameter, W, has the value 1.77E-02 at the tube outlet for the baseline case, meaning that so far as the tube wall material is concerned, recombination impedance is far more significant than diffusion. The concentration of tritium at the edge of the flow, calculated at the tube outlet, is only 2.86E-08 times that in the bulk. The reason it is so low is that the impedance to tritium permeation through the wall is very low in comparison with the difficulty of tritium diffusing through the bulk of the Flibe to the boundary layer at the edge of the flow. That low value is an early clue that permeation is not very strongly influenced by what is done with the tube surfaces. The last column of Table 1 lists the calculated partial pressure of the tritium in the Flibe at the wall as it exits the IHX. It is greater than the corresponding pressure at the inlet because the Flibe is cooler at the outlet, and the solubility is substantially lower. The Flibe concentration at the outlet is only 6% lower in the baseline case while the solubility is less than half as great, just due to the cooling of the Flibe, so the partial pressure of the tritium is increased.

The design parameters of the tube were varied in the subsequent calculations. First the recom-

bination coefficient was reduced to 10^{-5} of its reference value. This is one way of adding the effect of a permeation barrier such as an oxide layer. This change is reflected in the extremely low value of W, showing that recombination is even more significant than diffusion for this material condition. There was virtually no change in the permeation fraction, however.

The next change was to increase the flow velocity, u , by a factor of two. The tritium permeation fraction fell slightly while the other parameters listed remained about the same. The lower permeation fraction is the result of the tritium having less time to diffuse to the tube wall.

The wall thickness was increased from 1 mm to 5 mm. The permeation parameter, W, increased by the same proportion, meaning that the diffusive impedance in the wall has greater but still inconsequential significance. There was no change in the permeation fraction. Again, diffusion to the tube wall is much more limiting than either recombination or diffusion through the wall.

Increasing the tube radius, R, from 9 mm to 45 mm did have a strong effect on reducing permeation. This is a consequence of increasing the length through which the turbulent diffusion takes place. Doubling the tube length had the predictable effect of increasing the permeation fraction by almost a factor of two, and increasing the system temperatures by 100 K increased it as well. That is because of the increased turbulent diffusivity through the Flibe.

The final comparison in Table 1 shows the results of calculations with material properties of vanadium, tungsten and a composite consisting of 0.1 mm of tungsten overlaying the standard 1-mm thick stainless steel. The composite properties were evaluated from the properties of the individual layer materials using the formalism outlined by Abramov et al.⁹ that results in

$$D_{eff} = \frac{x^2}{\sum \frac{x_i^2}{D_i}} \quad (15)$$

for the effective value of diffusivity and

$$S_{eff} = \frac{\sum \frac{x_i^2}{D_i}}{x \sum \frac{x_i}{S_i D_i}} \quad (16)$$

for the effective solubility. Here x_i is the thickness of the respective layer, and x is the total thickness of the membrane. D_i and S_i are the diffusivity and solubility, respectively, for the layers. Though there was a change in the other parameters for each of these cases, there was no

effective change in the permeation fraction for any of them.

Additional calculations were performed in which the effects of viscosity were examined. Viscosity decreases with temperature in an Arrhenius fashion. When viscosity was increased to 40 times its normal value, the permeation fraction increased to 98% for the otherwise baseline calculation. The other cases presented in Table 1 when modified in the same way had the same general relationship to the modified baseline calculation as indicated in Table 1.

DISCUSSION

From the results just presented, it is apparent that the main impedance to loss of tritium through HYLIFE-II heat exchanger tubes is its transport through the flibe to the tube walls. The variations in problem parameters that had a favorable effect on the permeation fraction, specifically, increasing tube radius or fluid velocity or reducing the temperature of the IHX system, would all diminish the thermal performance of the system. On the other hand, changes made to the tubes themselves such as thickening tube walls or the application of a film or coating had virtually no effect. The relative invariance of the result to the initial partial pressure of tritium in the Flibe implies that the solubility of tritium in the salt has little influence on the permeation rate.

The notable increase in permeation with increased viscosity may be anticipated from the analytical model. In the opposite direction, a reduced viscosity means greater Reynolds numbers and an increased value of the dimensionless radius, r^+ , leading to an increase in the dimensionless concentration, $\langle c \rangle$. Reduced viscosity results in a much shallower gradient in concentration profiles in the bulk of the fluid. That has the effect of reducing radial transport to the walls and thus reducing tritium permeation through walls. Increasing the viscosity has the opposite effect.

Two different Flibe compositions are being considered for use in HYLIFE-II with different viscosities. The present calculated result is an argument in favor of the low-viscosity composition.

The tritium emission goal of 40 Ci/day is less than 10^{-5} of the tritium load coming from the blast chamber. Optimal design of the IHX probably means that the loss rate for tritium through that component will be a few percent such as calculated above. If the same general behavior could be obtained from the intermediate cooling system (NaBF_4), then the goal should be reachable. The vacuum disengager on the primary loop and the gas exchange column in the intermediate loop would each need to remove about 99% of the tritium entering them. However, confidence in that conclusion hinges on the permeation losses through the

intermediate loop. Transport properties for tritium in NaBF_4 are needed to make that estimation.

CONCLUSIONS

The combination of the Lewis analogy with notions of recombination-dissociation and diffusion limited permeation processes has resulted in a method for estimating tritium losses in heat exchanger tubes or other situations in which turbulent flow of a tritium bearing liquid is of concern. The computation is facilitated by the nondimensionalization of the permeation flux and use of the permeation parameter, W . That allows relatively straightforward estimation of the driving potential for tritium permeation through the tube walls.

Computations using that model indicated that the turbulent diffusion of tritium to the wall through the Flibe is the controlling factor in determining the fraction permeating. Because of that, there appears to be little that can be done by way of coatings or material changes in the wall to reduce permeation losses. On the other hand, it means that high-permeability materials such as vanadium may be considered for use in the IHX or similar systems for reasons of low activation or corrosion resistance with impunity from the tritium-loss perspective. Fluid viscosity is a strongly influential parameter, and lower viscosity means lower tritium permeation.

The calculations upon which these conclusions are based depend on the validity of the assumptions made. The estimates provided here need to be verified by experiment. Additionally, data on tritium transport in NaBF_4 are needed to allow estimates to be made of tritium losses in the intermediate heat transfer loop.

Further analysis is also needed to evaluate the performance of the vacuum disengager system in the primary loop and the gas equilibration system in the intermediate loop. Present estimates are that with reasonable performance in both of these systems, the tritium release goal of 40 Ci/day may be achieved.

REFERENCES

1. F. WAELBROECK et al., "Influence of Bulk and Surface Phenomena on the Hydrogen Permeation Through Metals," JUL-1966 (December 1984) 32, ISSN 0366-0885, Institut für Plasmaphysik, Association EURATOM-KFA, Kernforschungsanlage, Julich GmbH.
2. R. A. KERST and W. A. SWANSIGER, "Plasma Driven Permeation of Tritium in Fusion Reactors," *Journal of Nuclear Materials* 122 & 123, 1499 (1984).

3. B. L. DOYLE, "A Simple Theory for Maximum H Inventory and Release: A New Transport Parameter," *Journal of Nuclear Materials* 111 & 112, 628 (1982).
4. R. B. BYRD, W. E. STEWART, and E. N. LIGHTFOOT, *Transport Phenomena*, John Wiley & Sons, Inc., New York (1960) pp. 164, 187, and Sec. 20.3.
5. S. CANTOR et al., "Viscosity and Density in Molten LiF-BeF₂ Solutions," *J. Chem. Phys.* 50, 2874 (1969).
6. J. OISHI et al., "Tritium Recovery from Molten LiF-BeF Salt," *Fusion Engineering and Design*, 8, 317 (1989).
7. T. TERAJ et al., "Behavior of Hydrogen Isotopes in Molten Breeding Materials," Proc. Int. Symp. on Fusion Reactor Blanket and Fuel Cycle Technology, Tokai-mura, Ibaraki, Japan (October 27-29, 1986) p. 98.
8. M. I. BASKES, "A Calculation of the Surface Recombination Rate Constant for Hydrogen Isotopes on Metals," *Journal of Nuclear Materials*, 92, 32 (1980).
9. E. ABRAMOV et al., "Deuterium Permeation and Diffusion in High Purity Beryllium," *CFFTP-G-9013* (May, 1990) Canadian Fusion Fuels Technology Program, Mississauga, Ontario, Canada.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

END

DATE FILMED

01 / 31 / 91

