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TRITIUM PERMEATION IN FUSION REACTORS: INTOR

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ABSTRACT

Tritium permeation through the first wall of advanced fusion reactors is examined in this report. A fraction of the D-T which bombards the first wall as charge exchange neutral particles will permeate through the first wall and enter the coolant. Calculations of the steady state permeation rate for the U.S. INTOR Tokamak design result in values of ≤ 0.002 grams of tritium per day under the most favorable conditions. For unfavorable surface conditions the rate is ≥ 0.1 g/day. The magnitude of these permeation rates is critically dependent on the temperatures and surface conditions of the wall. The introduction of permeation barriers at the wall-coolant interface can significantly reduce permeation rates and hence may be desirable for reactor applications.

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INTRODUCTION

Tritium permeation through stainless steel first walls subjected to energetic tritium charge exchange neutral bombardment has been recently identified as a potentially serious problem area for advanced D-T reactors operating at elevated temperatures^[1,2]. High concentrations of tritium in the near surface region of the first wall can be achieved by implantation of the charge exchange neutral flux and a relatively slow recombination of these atoms into molecules at the plasma/first wall interface.

Because of this large concentration of mobile tritium near the inner (plasma) wall surface, a concentration gradient is established, causing tritium to diffuse into the bulk and eventually to the outer first wall surface where it can enter the first wall coolant.

Previous calculations by Wienhold, et al^[2] have shown that as much as 16g of tritium (1.6×10^5 Ci) per day could permeate through a stainless steel, INTOR-sized vessel operated at 600°C and bombarded with 10^{17} cm⁻² s⁻¹ flux to the first wall^[2]. However, the first wall temperatures used in their calculations are far in excess of those expected for INTOR operation, which are expected to be ~300°C on the inner (plasma) surface and ~100°C on the outer (coolant) surface. Calculations with a range of operating parameters appropriate for INTOR are summarized in this paper. In addition, the effect of coatings as permeation barriers on both the inner and outer wall surfaces is discussed.

THEORY

The calculations use the DIFFUSE^[3] numerical solution to the diffusion equation

$$\frac{\partial C}{\partial t} = -\frac{\partial J}{\partial x} + G \quad (1)$$

where C is the mobile tritium concentration, J is the bulk tritium flux, and G is the implant source term. Trapping of tritium at radiation-damage defects has been neglected in the calculations because of the relatively low binding energies involved^[4]. Deviations from Fickian diffusion due to the interactions of tritium with mobile defects^[5,6] are also not considered.

The surface boundary conditions are of the form:

$$\phi_s = K_r C_s^2 \quad (2)$$

where ϕ_s is the atom flux of tritium leaving the surface (as molecules), C_s is the near surface tritium concentration, and K_r is the T_2 molecular recombination rate constant. The appropriate values for K_r for the first wall surfaces of a fusion reactor are uncertain. Fig. 1 summarizes experimental values of K_r for stainless steels^[4]. The present calculations use the theory for K_r developed by Baskes^[7]:

$$K_r = 5.27 \times 10^{25} \alpha e^{(E_s - E_D)/KT} / C_0^{*2} \quad (3)$$

where E_s and E_D are the tritium solubility and diffusivity activation energies respectively, C_0^* is the Sieverts' Law constant, and α is the molecular

sticking coefficient on either the inner (facing the plasma) surface, or outer α_0 (facing the coolant) surface. Since the sticking coefficient depends on surface composition, cleanliness, etc., values of α_i from 0.5 to 5×10^{-5} are used at the inner surface. For the outer surface, α_0 is assumed to be a fraction of the inner surface value. The possibility that a different kinetics formalism applies to the outer surface is not considered.

Figure 1. A comparison of experimental data for k_r of stainless steel with the theory of Baskes^[7] for two values of the molecular sticking coefficient (α)^[4].

Since INTOR will be operated with a large incident power flux on the inner surface and water coolant on the outer surface of the first wall, a temperature gradient has been incorporated into the calculations. The gradient has two main consequences. First, the lower temperature at the outer surface leads to slower diffusion and outer surface release rates. Second, the temperature gradient can change the mobility of tritium due to the Soret effect^[8] whereby the tritium flux is controlled by the temperature gradient as well as the concentration gradient. The tritium flux inside the wall is therefore:

$$J = -D \left(\frac{\partial C}{\partial x} + \frac{CQ^*}{kT^2} \frac{\partial T}{\partial x} \right) \quad (4)$$

where D is the tritium diffusivity (pre-exponential D and activation energy E_D), and Q^* is the heat of transport. The first term gives the usual flux contribution due to a concentration gradient, while the second term accounts for the flux induced by the thermal gradient. For hydrogen in γ -Fe-Ni alloys and Ni, Q^* is ~ -0.65 eV^[9]. Hence, tritium will tend to diffuse to the hotter side of the first wall, lowering the permeation rate.

Table I summarizes parameters employed in the calculations. A linear temperature gradient is used for the first wall, with the inner surface at 300°C and the outer surface at 100°C^[1], except for sensitivity studies.

TABLE I

Material Parameters

Wall Thickness	1.34 cm
D_O	$0.085 \text{ cm}^2\text{-sec}^{-1}$
E_D	0.61 eV
E_S	0.091 eV
C_O^*	$7.65 \times 10^{19} \text{ atoms cm}^{-3}\text{-atm}^{-\frac{1}{2}}$
Q^*	-0.065 eV

Tritium Charge Exchange Neutral Flux Parameters

Flux	$3.3 \times 10^{16} \text{ D\&T cm}^{-2}\text{-sec}^{-1}$
Energy distribution	200 eV Maxwellian
Angular distribution	cosine
Discharge time	200 sec
Duty cycle	.816

RESULTS AND DISCUSSION

Figure 2 shows the steady state permeation rates obtained for the standard INTOR conditions, with the inner surface molecular sticking coefficient equal to 0.5 ("clean" surface) or 5×10^{-5} ("dirty" surface) and the outer surface sticking coefficient a fraction, from unity to 10^{-4} , of the inner surface value. The highest tritium permeation rate (>0.1 g/day) is obtained for a "dirty" inner surface ($\alpha_i = 5 \times 10^{-5}$), and relatively high recombination ($\alpha_o/\alpha_i \geq 1$) at the outer surface. As $\alpha_o/\alpha_i \rightarrow \infty$, the tritium permeation rate approaches 5.5×10^{-3} g/day for a "clean" inner surface and 0.55 g/day for a "dirty" inner surface. The condition $\alpha_o/\alpha_i \rightarrow \infty$ is equivalent to a zero outer surface tritium concentration and yields the maximum permeation rate independent of the outer surface kinetics. Under the ideal conditions of a "clean" inner surface ($\alpha_i = 0.5$) and slow recombination at the coolant ($\alpha_o/\alpha_i \leq 10^{-4}$), tritium permeation is reduced to <0.002 g/day. Note that for a "clean" inner surface the condition of the outer surface is relatively unimportant for $\alpha_o/\alpha_i \geq 10^{-4}$.

The importance of using the correct temperature and temperature gradient are shown in Table II. There is over two orders of magnitude difference between an INTOR wall with a temperature gradient (300°C inner surface/100°C outer surface) and one held uniformly at 300°C. The Soret effect contributes only a factor of two to the difference. The main contribution of the temperature gradient is to reduce the temperature and hence lower the diffusivity near the outer surface of the wall, and to lower the outer surface recombination constant. For a wall operated with the same temperature gradient, but with

TRITIUM PERMEATION

Figure 2. Calculated Tritium Permeation Rates for an INTOR First Wall Under standard operating conditions. Both "clean" ($\alpha_i = 0.5$) and "dirty" ($\alpha_i = 5 \times 10^{-5}$) inner wall surface conditions are considered.

TABLE II

Temperature		Soret Effect	Steady State Tritium Permeation (g/day)
Inner	Outer		
300°C	100°C	Yes	0.0047
300°C	100°C	No	0.0086
300°C	300°C	--	0.78
500°C	300°C	Yes	0.61
500°C	500°C	--	5.2

A sticking coefficient of $\alpha = 0.5$ was used for both surfaces.

the inner surface at 500°C and outer surface at 300°C (DEMO-like conditions)^[10] the permeation rate is two orders of magnitude higher than for INTOR-like conditions. Finally, for the wall held at the unrealistic case of a uniform 500°C temperature, the tritium permeation rate is 5 g/day.

It is likely that a low Z coating, e.g., C, TiC, TiB₂, etc., on the inner wall may reduce the tritium permeation rate. A number of implantation studies^[11-14] have shown that such coatings trap essentially all implanted hydrogen atoms until a critical hydrogen lattice concentration is reached. Any additional implanted hydrogen is then released at the implant surface. A second proposal for the reduction of permeation is the use of thin metallic coatings (e.g., Pd) on the inner surface to enhance recombination^[15-16].

The permeation rate can also be lowered significantly by the use of permeation barriers at the outer surface. Table III summarizes the results of calculations for a bare stainless steel first wall (with a 300°C to 100°C temperature gradient, $\alpha_i = \alpha_o = 5 \times 10^{-5}$), compared to similar first walls with 10 μ m thick coatings of Au, W, Al₂O₃ or Si on the outer surface. Literature permeation rates through these coatings were extrapolated to 100°C. It is also possible that naturally occurring oxides on the stainless steel-water coolant interface can act to reduce permeation. Clearly, the tritium permeation rates can be made acceptably low for the INTOR design by the use of these permeation barriers. Of course, the viability of coatings or oxides on the outer as well as inner surface with respect to thermal cycling, coolant compatibility and plasma erosion/redeposition, etc., remains to be investigated.

TABLE III

First Wall	Tritium Permeation (g/day)
Stainless Steel	0.42
SS/Au	9.8×10^{-6}
SS/W	3.35×10^{-8}
SS/ Al_2O_3	1.54×10^{-16}
SS/Si	3.46×10^{-20}

Steady state tritium permeation rates for stainless steel walls operated with a 300°C/100°C temperature gradient. All coatings are 10 μm thick.

CONCLUSIONS

For a stainless steel wall subjected to an energetic tritium charge exchange neutral flux, the tritium permeation rate through the first wall can be quite large. The magnitude of the effect is critically dependent on the temperatures and surface conditions assumed for the wall. For INTOR-like conditions, a steady state rate of $\sim 10^{-3}$ g/day is possible under optimum conditions while for a first wall at more elevated temperatures (such as DEMO) the tritium permeation rate can easily exceed 0.5 g/day. These tritium permeation rates can be reduced significantly by the use of permeation barriers at the first wall-plasma or first wall-coolant interfaces.

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