

Analysis of first and second order surface processes in fusion solid breeders

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Surface reactions in tritium release from fusion ceramic breeders were investigated. Analyses of the tritium inventory were made under steady state conditions using first and second order adsorption/desorption processes on the surface. The resulting equations were used to calculate the grain and surface inventories for different temperatures ranging from 350°C to 900°C, and different H₂ concentrations ranging from 0% to 10%, using data from Li₂O samples resulted from the experiments named TTTEEx. Comparison between the analysis and the experimental data leads to the conclusion that in order to study the behavior of tritium inside the ceramic breeders, second order surface processes must be used in modeling and calculating the tritium release and inventory. Although this analysis used data from Li₂O samples, there would be no need to repeat the calculations using other materials. The results of this study apply to most ceramic breeders, since the tritium exhibits the same behavior when other materials are used.

تمت في هذا البحث دراسة تفاعلات السطح لتحرير التريتيوم من مولدات الاندماج الخزفية. كما تم تحليل جرد التريتيوم في الحالات الثابتة مع الزمن، وذلك باستخدام عمليات أدمصاص و مخرج من الدرجة الأولى و الثانية على السطح. استعملت المعادلات الناتجة لحساب الجرد في الحبيبات و على السطح لدرجات حرارة مختلفة بين 350 و 900 درجة مئوية، و تركيز هيدروجين مختلف بين 0% و 10%، باستخدام البيانات الخاصة بعينات أكسيد الليثيوم المستخدمة في مجموعة تجارب TTTEEx. أدت المقارنة بين التحليل النظري و النتائج التجريبية إلى أنه حتى يمكن دراسة سلوك التريتيوم داخل المولدات الخزفية، يجب استخدام عمليات سطح من الدرجة الثانية في النماذج الموضوعية لحساب تحرير و جرد التريتيوم. بالرغم من استخدام البيانات الخاصة بعينات أكسيد الليثيوم، فإنه من الممكن تطبيق نفس النتيجة على أغلبية المولدات الخزفية، وذلك لأن سلوك التريتيوم متشابه في معظم المولدات الخزفية.

Keywords: Solid breeders, Adsorption, Desorption, Surface coverage, Tritium inventory.

1. Introduction

Precise evaluation of tritium inventory in the fusion blanket is essential for the design of the fusion reactor fuel cycle. Tritium transport and release from the solid breeding material are complex processes that include both diffusion as well as surface reactions (such as adsorption, desorption, and dissolution processes)[1]. The rate-limiting process is dependent on the operating conditions, such as temperature, purge gas composition, and microstructure parameters such as grain size and porosity distribution [2]

It has been reported that at high temperature the rate-determining processes of tritium release are surface process [3]. Also, at high temperatures, the surface inventory was found to be larger than that due to diffusion in the absence of H₂ or water vapor in the purge gas [4].

In most studies, adsorption/desorption processes are considered to be first-order reactions. However, in some experiments it was found that they can be of a second order [5]. Hence, there is a need to know how the modeling results will differ in each case, as well as find out whether both first and second order reactions can be used to simulate the phenomena associated with tritium transport in ceramic breeders.

In this paper, the effect of the order of surface reactions was investigated. Both the tritium inventories in the grain and on the surface were examined as a function of temperature, in the presence and absence of H-atoms in the purge gas. Data from the TTTEEx experiments [6-10] were used to calculate the surface and grain inventories in different conditions. The phenomena studied in this paper are the decrease of tritium inventory with increasing temperature, and its

decrease with the addition of H₂ to the sweep gas. Steady state was assumed to simplify the equations to be solved analytically and not numerically as in the case of transient conditions. This gives a better understanding of the phenomena involved in the tritium release and retention. It should be noted that using either steady state or transient conditions has no effect on the order of surface reactions, which is the focus of this paper.

2. The surface fluxes

A schematic diagram showing the fluxes due to different processes on the surface is shown in Figure 1. There are mainly 4 fluxes going to and from the surface [11]:

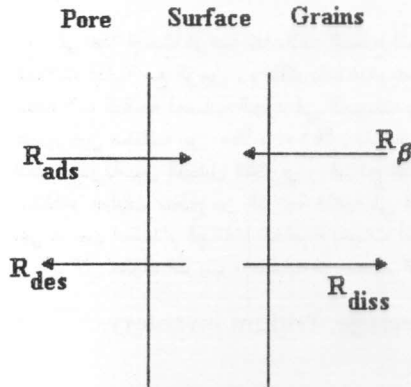


Fig. 1. Schematic diagram showing the different surface fluxes.

1. A flux, R_{ads} (at/m².s), going from the pore to the surface. For first order adsorption [11],

$$R_{ads} = \frac{\sigma \cdot z}{\sqrt{8 \times 10^{-3} \pi}} \sqrt{\frac{RT}{M}} C_p (1 - \theta) \exp(-E_{ads} / RT). \quad (1)$$

Where C_p is the concentration of the adsorbed species in the pore (at/m³), θ is the total surface coverage (i.e. the fraction of filled surface sites). The term $(1 - \theta)$ represents the number of empty sites available for adsorption. E_{ads} is the activation energy of adsorption (J/mol), R is the universal gas constant (J/mol.K), T is the temperature (K), σ is the sticking coefficient, z is the number of

sites adjacent to each atom, and M is the molecular weight. For second order adsorption [12],

$$R_{ads} = \frac{\sigma \cdot z}{\sqrt{8 \times 10^{-3} \pi}} \sqrt{\frac{RT}{M}} C_p (1 - \theta)^2 \exp(-2E_{ads} / RT). \quad (2)$$

Note that the squaring of $(1 - \theta)$ and the doubling of E_{ads} stems from the fact that two atoms are required to adsorb in order to form H₂, HT, or T₂ [13].

2. A flux, R_{des} (at/m².s), going from the surface to the pore. For first order desorption [13],

$$R_{des} = \frac{RN_s z T}{A_v h} \theta \exp(-E_{des} / RT). \quad (3)$$

Where E_{des} is the activation energy of desorption (J/mol), θ is the surface coverage of the desorbing species, N_s is the number of sites on the surface of the grain (at/m²), A_v is Avogadro's number (mol⁻¹), and h is Planck's constant (J.s). For second order desorption [12],

$$R_{des} = \frac{RN_s z T}{A_v h} \theta^2 \exp(-2E_{des} / RT). \quad (4)$$

3. A flux, R_{diss} (at/m².s), going from the surface back to the grain,

$$R_{diss} = \frac{RN_s z T}{A_v h} \theta \exp(-E_{diss} / RT). \quad (5)$$

Where, E_{diss} is the activation energy of dissolution (J/mol).

4. A flux, R_β (at/m².s), going to the surface from the bulk [11],

$$R_\beta = \frac{3 \times 10^{13}}{r_g S_{BET} \rho \sqrt{N_s}} C_b (1 - \theta) \exp(-E_\beta / RT). \quad (6)$$

Where, C_b is the tritium concentration in the bulk just below the surface (at/m³), E_β is the

activation energy for adsorption from the bulk to the surface (J/mol), S_{BET} is the specific surface area (m²/kg), r_g is the grain radius (m), and ρ is the theoretical density (kg/m³). The four energies, E_{ads} , E_{des} , E_{diss} , and E_β are related through the activation energy of solution, E_s , and the heat of adsorption, Q [12]:

$$E_s = E_{ads} - E_{des} + E_{diss} - E_\beta, \quad (7)$$

$$Q = E_{des} - E_{ads}. \quad (8)$$

Eqs. (1), (3), (5), and (6) can be rewritten as:

$$R_{ads} = K_{ads} (1 - \theta), \quad (9)$$

$$R_{des} = K_{des} \theta, \quad (10)$$

$$R_{diss} = K_{diss} \theta, \quad (11)$$

$$R_\beta = K_\beta (1 - \theta) C_b, \quad (12)$$

$$K_{ads} = \frac{\sigma z}{\sqrt{8 \times 10^{-3}} \pi} \sqrt{\frac{RT}{M}} C_p \exp(-E_{ads} / RT), \quad (13)$$

$$K_{des} = \frac{RN_s z T}{A_v h} \exp(-E_{des} / RT), \quad (14)$$

$$K_{diss} = \frac{RN_s z T}{A_v h} \exp(-E_{diss} / RT), \quad (15)$$

$$K_\beta = \frac{3 \times 10^{13}}{r_g S_{BET} \rho \sqrt{N_s}} \exp(-E_\beta / RT). \quad (16)$$

The equations that describe the surface fluxes, Eqs. (9) and (12), are used to calculate the tritium surface coverage, θ_T , and the tritium grain concentration, C_b . In the case of absence of protium in the purge gas, the total surface coverage (θ) in the above equations is equal to the tritium surface coverage (θ_T). When protium is added, the fraction of filled sites (θ) will be equal to ($\theta_T + \theta_H$). The tritium surface inventory, I_s (g), is related to the surface coverage by [13]

$$I_s = \frac{\theta_T N_s S_{BET} \rho V_{br} M (1 - \epsilon)}{A_v}, \quad (17)$$

$$I_g = \left(C_b + \frac{gr_g^2}{15D} \right) \frac{V_{br} M (1 - \epsilon)}{A_v}. \quad (18)$$

Where V_{br} is the breeder volume (m³) and ϵ is its porosity. The grain inventory, I_g (g), can be obtained from C_b , D is the diffusion coefficient of tritium in the grains (m²/s). In this case g is the rate of tritium generation per unit volume of the breeder (at/m³.s). The total tritium inventory in the breeder is equal to

$$I_{tot} = I_s + I_g. \quad (19)$$

Case 1: First order adsorption/desorption

For the case of steady state tritium generation and release, the net flux coming out of the pores is equal to the tritium generated inside the grains. Therefore,

$$R_{des,T} - R_{ads,T} = g. \quad (20)$$

Where, g is the effective rate of tritium generation per unit surface area (at/m².s).

If no H₂ is present in the purge gas the surface will be covered with only tritium. Therefore, the total surface coverage θ will be equal to the tritium coverage θ_T . Rewriting Eq. (20)

$$K_{des} \theta_T - K_{ads,T} (1 - \theta_T) = g \quad (21)$$

Note that K_{des} does not depend on whether the flux is that of tritium or hydrogen, whereas K_{ads} is isotope-dependent through both the molecular weight and the pore concentration of the adsorbed gas. Hence, the subscript (T) will denote the tritium while the subscript (H) will denote the protium.

Solving eq. (21) for the tritium surface coverage we get:

$$\theta_T = \frac{g + K_{ads,T}}{K_{des} + K_{ads,T}}. \quad (22)$$

Since the net flux leaving the grain must also be equal to the generated tritium, we can write:

$$R_\beta - R_{diss} = g, \quad (23)$$

$$K_{\beta}C_b(1-\theta_T) - K_{diss} \theta_T = g. \quad (24)$$

Substituting for θ_T with Eq. (22), we get

$$C_b = \frac{g(K_{des} + K_{ads,T} + K_{diss}) + K_{diss} K_{ads,T}}{K_{\beta}(K_{des} - g)}. \quad (25)$$

In the presence of H_2 in the purge gas, the equations become more complicated due to the fact that the surface sites will be covered with both protium and tritium, i.e.

$$\theta_{tot} = \theta_T + \theta_H. \quad (26)$$

Thus an additional equation is required to account for θ_H . This can be obtained from equating both the desorption and adsorption fluxes of protium at steady state (since no protium is generated in the breeder)

$$R_{des,H} = R_{ads,H}, \quad (27)$$

$$K_{des} \theta_H = K_{ads,H} (1 - \theta_{tot}). \quad (28)$$

Again for tritium, solving Eq. (20) we get:

$$K_{des} \theta_T = K_{ads,T} (1 - \theta_{tot}) = g. \quad (29)$$

Solving Eqs. (26), (28), and (29) we obtain two equations that describe the surface coverage of tritium and protium in terms of the different flux coefficients:

$$\theta_T = \frac{g(K_{des} + K_{ads,H}) + K_{des} K_{ads,T}}{K_{des}(K_{des} + K_{ads,T} + K_{ads,H})}, \quad (30)$$

$$\theta_H = \frac{K_{ads,H}(K_{des} - g)}{K_{des}(K_{des} + K_{ads,T} + K_{ads,H})}, \quad (31)$$

$$K_{\beta}C_b(1 - \theta_{tot}) - K_{diss}\theta_T = g. \quad (32)$$

Equation (23) is used to obtain the tritium grain concentration

$$C_b = \frac{K_{ads,T}(g + K_{diss}\theta_T)}{K_{\beta}(K_{des}\theta_T - g)}. \quad (33)$$

Case 2: Second order adsorption/ desorption

For second order surface processes, the same basic equations are used. However, the term $(1 - \theta)$ in the adsorption flux is squared together with the term θ in the desorption flux. Also in K_{des} and K_{ads} , the activation energies are doubled. Therefore, for steady state conditions with no H_2 in the purge gas, solving Equation 20 gives:

$$K_{des} \theta_T^2 - K_{ads,T} (1 - \theta_T)^2 = g. \quad (34)$$

Solving for θ_T we get:

$$\theta_T = \frac{-K_{ads,T} + \sqrt{K_{ads,T}^2 + (g + K_{ads,T})(K_{des} - K_{ads,T})}}{K_{des} - K_{ads,T}}. \quad (35)$$

The tritium bulk concentration can be obtained from Eq. (23),

$$C_b = \frac{g + K_{diss}\theta_T}{K_{\beta}(1 - \theta_T)}. \quad (36)$$

For the addition of protium to the purge gas, there will be two different gases containing protium in the system, namely, H_2 and HT. By equating the different protium adsorbing and desorbing fluxes, we get:

$$K_{des} \theta_{tot} \theta_H = K_{ads,H} (1 - \theta_{tot})^2. \quad (37)$$

The term $\theta_{tot}\theta_H$ is due to the addition of a term containing $\theta_H\theta_H$, of the desorbing H_2 flux and a term containing $\theta_T\theta_H$ of the desorbing HT flux [13].

Similarly, for tritium, considering that there are two tritium containing gases; T_2 and HT, we get:

$$K_{des} \theta_{tot} \theta_T - K_{ads,T} (1 - \theta_{tot})^2 = g. \quad (38)$$

Solving Eqs. (37), (38), and (26), we get a relation between the total surface coverage and that of tritium:

$$\theta_{\text{tot}} = \frac{K_{\text{tot}} - \sqrt{K_{\text{tot}}^2 - (g + K_{\text{tot}})(K_{\text{tot}} - K_{\text{des}})}}{K_{\text{tot}} - K_{\text{des}}}, \quad (39)$$

$$K_{\text{tot}} = K_{\text{ads,T}} + K_{\text{ads,H}}, \quad (40)$$

$$\theta_{\text{T}} = \frac{g + K_{\text{ads,T}}(1 - \theta_{\text{tot}})^2}{K_{\text{des}}\theta_{\text{tot}}}. \quad (41)$$

The tritium concentration in the grain is then obtained from Eq. (32).

3. Examination of tritium transport phenomena

The tritium release behavior from Li_2O and LiAlO_2 was studied in a set of experiments named as TTEx [6-10]. In these experiments, the samples were irradiated, and the amount of tritium released was measured at different temperatures and purge gas compositions. Although the experiments were made using Li_2O and LiAlO_2 , only Li_2O data are used in the analysis because: (1) it is one of the major candidates for the reactor blanket [13], and, (2) experimental data are available for Li_2O properties such as diffusion, desorption, adsorption and solubility [14]. This data is not readily available for other ceramic breeders.

Some of the sample and purge gas characteristics are summarized in Table 1. Those values are used to examine the surface and grain inventories and their relation with the temperature and H_2 concentration in the purge gas, for both first and second order surface reactions. The values in Table 1 were used to calculate the flux coefficients K_{des} , K_{ads} , K_{β} and K_{diss} . The tritium concentration $C_{p,T}$ was calculated from [12]:

$$C_{p,T} = \frac{gV_{\text{br}}(1 - \epsilon)}{Q}. \quad (42)$$

Where, Q is the purge gas flow rate (m^3/s).

3.1 Effect of temperature

Figure 2 shows the change in surface, grain and total tritium inventories with

temperature for first order reactions in the absence of H_2 in the purge gas. As expected, the inventory decreases with increasing temperature, since all of the processes are temperature-enhanced. I_{tot} decreases by 4 orders of magnitude between 350°C and 900°C . Although the total tritium inventory is almost equal to that in the grain, it is important to include surface processes when calculating the inventory. This is because the surface fluxes act as a barrier that slows down the tritium release, hence increasing the tritium inventory.

Table 1 Summary of the used TTEx experimental data [6-10].

Sample mass (g)	0.79
Porosity	0.22
Specific surface area (m^2/kg)	0.09
Sample volume (m^3)	5.32×10^{-4}
Grain radius (μm)	1.9
Rate of tritium generation (at/m^3)	2.83×10^{12}
Helium pressure (atm)	1
Flow rate (m^3/s)	2×10^{-6}

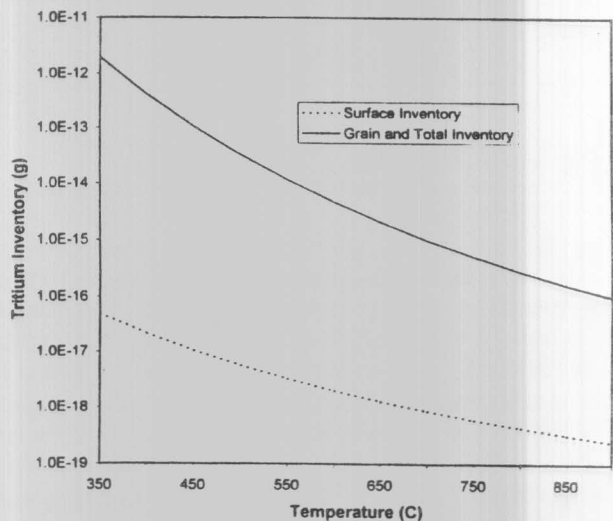


Fig. 2. Effect of temperature on tritium inventory for first order reactions in the absence of H_2 in the purge gas.

In Fig. 3 the change in inventory with temperature for second order adsorption/desorption processes in the absence of H_2 in the purge gas is shown. The

inventory is again found to decrease with increasing temperature. However, most of the inventory in this case is located on the surface, which is consistent with the findings of Nishikawa et al. [3] that in the absence of H₂ in the purge gas the surface inventory can be larger than that in the grain. This can be explained by noting that second order desorption is more difficult than first order desorption, because two atoms are required to combine and desorb together [11]. This makes it harder for tritium atoms to leave the surface, which causes an increase in the surface inventory.

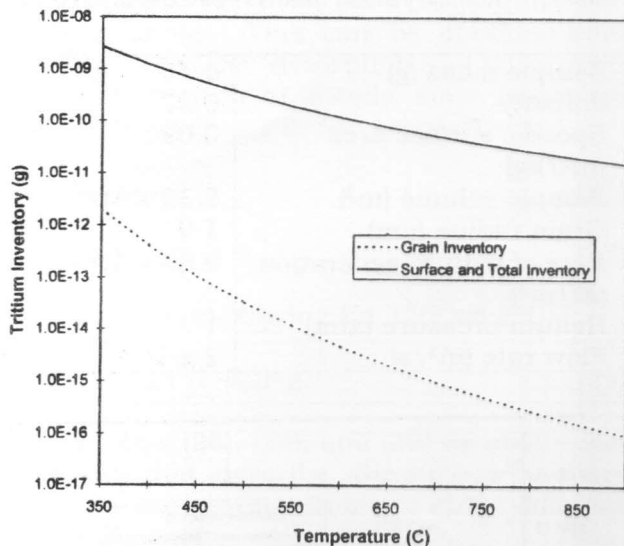


Fig. 3. Effect of temperature on tritium inventory for second order reactions in the absence of H₂ in the purge gas.

It can be seen from Figures 2 and 3 that the total inventory for the first order processes is much less than that in the second order case (about 2-3 orders of magnitude lower). For Li₂O samples, the tritium inventory in the TTTE experiments was between 10⁻⁸-10⁻¹⁰g at 410°C [9]. Comparison between the experimental and analytical results suggests that the surface processes are indeed second order and not first order processes.

Figure 4 shows the change in surface, grain and total tritium inventories with temperature for first order reactions in the presence of 1% H₂ in the purge gas. The figure is almost identical to Fig. 2, suggesting a negligible influence of adding H₂ to the purge

gas. This is in contrast to the experimental finding [6].

Figure 5 shows the effect of temperature on the tritium inventory for second order reactions in the presence of 1% H₂ in the purge gas. The surface inventory is less than in the case without protium (Figure 3). For example, at 600°C I_s decreased from 1.2 x 10⁹g to 6.3 x 10⁻¹⁴g when 1% H₂ was added to the purge gas. This is due to the swamping effect of the H₂ that competes with the tritium on the surface [10]. The surface inventory decreases with temperature, whereas the grain inventory decreases with temperature until about 600°C then starts to increase slightly. In order to explain this behavior, Eq. (32) can be rewritten as,

$$C_b = \frac{g}{K_\beta(1-\theta_{tot})} + \frac{K_{diss}\theta_T}{K_\beta(1-\theta_{tot})} \quad (43)$$

The first term in the RHS of Eq. (43) decreases with temperature, whereas the second term increases with temperature. At high temperatures the second term starts to dominate, causing a net increase in the tritium grain concentration and hence inventory.

The total inventory decreases with temperature before 600°C, as both I_s and I_g decrease with increasing temperature. After 600°C, the combination of a decreasing I_s and an increasing I_g produce a total inventory that does not change with temperature.

3.2. Effect of H₂ concentration

Figure 6 shows the effect of increasing the H₂ concentration in the purge gas on the tritium inventory for first order reactions at 650°C. The figure shows that the addition of H₂ to the helium purge gas has no effect on I_s, I_g, or the total inventory.

In all cases, θ_T was very small (≈ 4 x 10⁻¹³) and stayed constant for all H₂ concentration. Again this is in contrast with the experiment findings that adding H₂ to the He sweep gas decreases the tritium inventory [16,17].

In Fig. 7, the effect of H₂ concentration on the tritium inventory for second order surface reactions at 650°C is shown. Adding H₂ to the

purge gas decreases the inventory. The grain, surface, and total inventories decreased until the H₂ concentration reached about 1%. After this concentration, an increase in the amount of H₂ did not change the total tritium inventory. This was reported by Terai et al. [18] who found that the tritium residence time decreases with increasing concentration of H₂ in helium sweep gas up to a limit concentration above which it becomes constant.

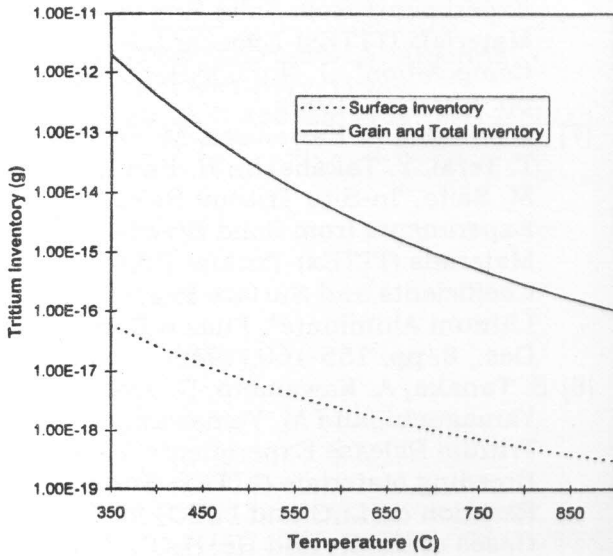


Fig. 4. Effect of temperature on tritium inventory for first order reactions in a sweep gas of He + 1% H₂.

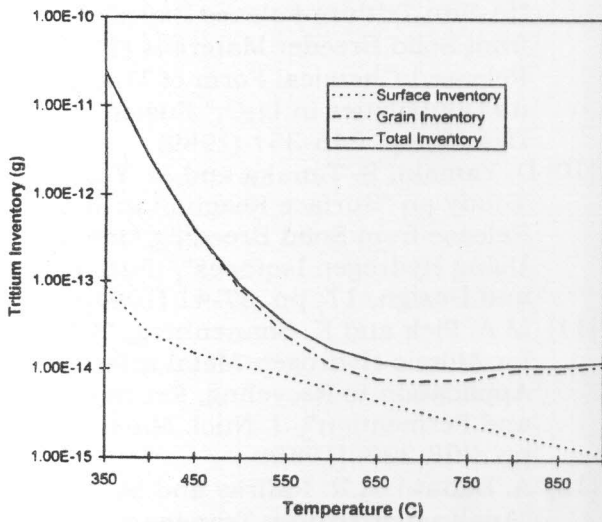


Fig. 5. Effect of temperature on tritium inventory for second order reactions in a sweep gas of He + 1% H₂ at 650°C.

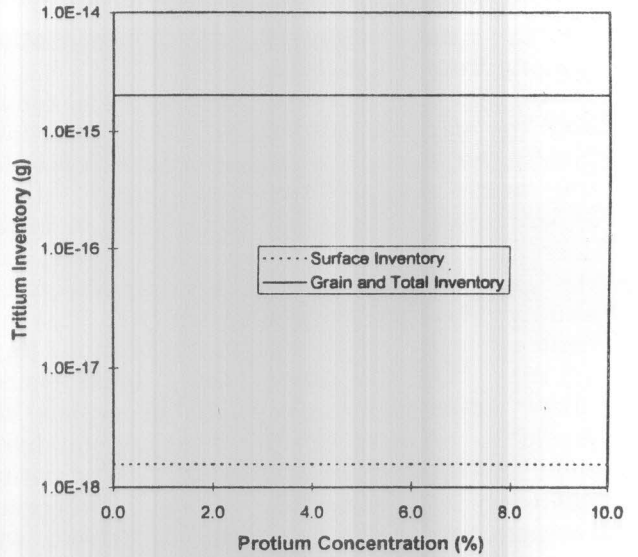


Fig. 6. Effect of H₂ concentration in the purge gas on the tritium inventory at 650°C for first order reactions.

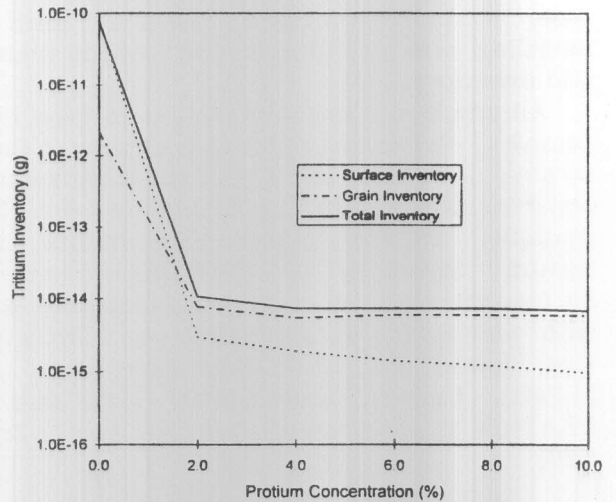


Fig. 7. Effect of H₂ concentration in the purge gas on the tritium inventory at 650°C for second order reactions.

4. Conclusion

Analyses of the tritium inventory in fusion ceramic breeders were made under steady state conditions, using first and second order adsorption/desorption processes on the surface. The resulting equations were used to calculate the grain and surface inventories for

different temperatures ranging from 350°C to 900°C, and different H₂ concentrations ranging from 0% to 10%.

Comparison between the analytical results and the experimental data shows that using first order surface reactions cannot reproduce the same tritium behavior found in the experiments. Although the tritium inventory decreases with increasing temperature, as in the experimental results, it is not affected by adding H₂ to the helium sweep gas. This is in contrast with the experimental findings [6, 8-10,16]. On the other hand, using second order adsorption/desorption processes was found to produce results that are consistent with the experimental results. The tritium inventory decreased with increasing temperature. The inventory decreased with increasing H₂ concentration up to a limit then remained constant for large values of H₂ concentration (>1%). This leads to the conclusion that in order to study the behavior of tritium inside the ceramic breeders, second order surface processes must be used in modeling and calculating the tritium release and inventory.

Although the analysis used data from Li₂O samples, the same conclusion applies to most ceramic breeders, since the same behavior is observed when other materials are used. For example, in the TTTEEx experiments, the tritium inventory in the LiAlO₂ samples decreased with increasing temperature and with adding H₂ to the purge gas. The same was found in EXOTIC-6 experiment for Li₂ZrO₃, Li₄SiO₄, and LiAlO₂ [19] and in BEATRIX II experiment for Li₂O and Li₂ZrO₃ [20].

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