ELSEVIER
Fusion Engineering and Design 28 (1995) 278–285
Fusion Engineering and Design

Analysis of tritium extraction from liquid lithium by permeation window and solid gettering processes

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Abstract

Tritium recovery from liquid lithium at low concentrations is an important problem in the study of liquid metal breeder-blankets in a fusion reactor. In this paper, we focus on the analysis of tritium extraction by permeation window and gettering processes. A time-dependent mass conservation equation for tritium in the liquid lithium and a diffusion equation for tritium transport in the permeation tube were solved simultaneously. The effects of the diffusivity in the permeation tube were discussed. In the gettering process, it is important to characterize the tritium breakthrough curves as a function of the getter material characteristics, operating conditions, equilibrium adsorption data and mass transfer coefficients. The analysis approach utilized in the present paper involves solving the momentum equation of the liquid lithium and the mass conservation equation of the tritium in the packed bed column, coupled with the diffusion equation of the tritium in a single particle. It was found that the tritium breakthrough curve at the near-wall region is different from that at the center region in the getter bed.

1. Introduction

Tritium recovery from liquid lithium at low concentrations is an important problem in the study of liquid metal breeder-blankets in a fusion reactor. Previous studies have identified tritium recovery methods such as molten salt extraction [1,2], gettering recovery [3,4], permeation windows [5,6], combined gettering and molten salt [7], and vacuum distillation [8]. In this paper, we focus on the numerical studies of tritium extraction by permeation window and gettering processes. These studies include dynamic tritium concentration variation along the flow direction; tritium inventory distributions in the permeation window and the getter bed; along with the effect of dispersion on the extraction efficiency. Using a model description makes it possible to determine the functional dependence, and provides an insight into the interrelationships of the various operating conditions and material properties which may affect the behavior of tritium in the material. Clearly, reliable material properties (such as diffusivity, solubility, etc.) are essential for realistic evaluations.

A typical permeation window resembles a counter-current shell-and-tube heat exchanger. Liquid lithium flows through the zirconium tube, while a purge stream containing oxygen flows along the shell side which is coated with palladium. Previous studies [5,6] have

SSDI 0920-3796(94)00103-1

shown that the rate of tritium extraction from the liquid lithium is dominated by the surface reaction process. Hsu and Buxbaum discussed using a palladium-coated zirconium membrane for permeation extraction. From ref. [6], the tritium transport resistance in liquid lithium can be taken as negligible. Tritium oxidation at the palladium surface limits tritium transport for some of the lithium-based windows; diffusion in the permeation membrane limits transport for others. However, because this conjugate phenomenon combines convection, diffusion and chemical reactions in a complicated manner, it is important to consider the effects of diffusion and convective mass transfer on tritium transport as the flow proceeds downstream. In the present analysis, a time-dependent mass conservation equation for tritium in the liquid lithium and a diffusion equation for tritium transport in the permeation tube were solved simultaneously, in order to investigate the effect of the tritium diffusivity in the permeation tube.

Tritium extraction using the gettering recovery process is based on the development of effluent concentration time curves, which are usually referred to as breakthrough curves. The effect of a non-uniform flow distribution of a fixed bed adsorber was investigated numerically by Vortmeyer and Michael [9]. This study has shown that the breakthrough curve depends greatly on the velocity profile while in the packed bed column. It is also important to characterize the tritium breakthrough curves as a function of the getter material characteristics, operating conditions, equilibrium adsorption data and mass transfer coefficients.

In the present paper, we focus on tritium extraction from liquid lithium using an yttrium getter bed. It was shown that the tritium breakthrough curve depends not only on the velocity profile but also on the velocity magnitude and equilibrium adsorption data. The analysis approach involves solving the momentum equation for the liquid lithium and the mass conservation equation for the tritium in the packed bed column, combined with the diffusion equation of the tritium in a single particle.

2. Permeation window

2.1. Numerical model and basic equation

To obtain the transient tritium concentration distribution in the permeation window along the axial and radial directions, the two-dimensional diffusion equation and one-dimensional mass conservation equation are solved numerically. The two-dimensional diffusion equation using cylindrical coordinates is

$$\frac{\partial C_S}{\partial t} = D_{TM}\left\{\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C_S}{\partial r}\right) + \frac{\partial^2 C_S}{\partial z^2}\right\} \tag{1}$$

where C_S is the tritium molar density in the solid phase and D_{TM} is the tritium diffusivity in the permeation tube. Some of the tritium diffusivity in the metal is described in Ref. [10]. For example, in zirconium, niobium and vanadium the tritium diffusivities at 450°C are 4.3×10^{-10}, 5.3×10^{-9}, 8.5×10^{-9} $m^2\,s^{-1}$ respectively.

The one-dimensional mass conservation equation is

$$\frac{\partial C_A}{\partial t} + \frac{\partial (C_A u)}{\partial z} = D_{TL}\frac{\partial^2 C_A}{\partial z^2} - \alpha_f \frac{L_h}{A_f}(C_A - C_{Ai}) \tag{2}$$

where C_A is the tritium molar density in the liquid phase and D_{TL} is the tritium diffusivity in liquid lithium.

Though there are many literature values for the tritium diffusivity in liquid lithium, we used the following equation [11].

$$\log D'_{TL} = -9.038 + 1.737 \log T - 110/T \quad (cm^2\,s^{-1}) \tag{3}$$

It should be noted that recent experimental results [12] suggested a slightly different temperature dependence on the value of the tritium diffusivity as compared with those used in the present analysis. L_h is the length of wet periphery and A_f is the flow area of the tube. The flow velocity in the tube (u) is assumed to be constant.

To investigate the effects of diffusion in the permeation tube and convective mass transfer on tritium transport as the flow proceeds downstream, the boundary condition on the outer surface of the tube is assumed to be $C_S = 0$. In other words, the tritium that reaches the outer palladium surface can be removed immediately by oxidation. The boundary condition on the inner surface of the tube is given by

$$-D_{TZ}\frac{C_S - C_{Si}}{\Delta r} = \alpha_f (C_A - C_{Ai}) \tag{4}$$

where C_{Si} is the interfacial molar density in the solid phase. It is assumed that equilibrium is not reached in the tube surface for higher flow velocities. Therefore, C_{Si} is equal to the interfacial molar density (C_{Ai}) in the fluid phase.

The mass transfer coefficient in a circular tube (α_f) is given by an empirical mass transfer coefficient, which is obtained by [13]

$$\mathrm{Sh} = \frac{\alpha_f d}{D_{TL}} = 0.023\,\mathrm{Re}^{0.83}\mathrm{Sc}^{1/3} \quad (2000 < \mathrm{Re} < 70\,000;\ 0.6 < \mathrm{Sc} < 2500) \tag{5}$$

where Sh is the Sherwood number, d is the inner diameter of the tube, $\mathrm{Re}(=ud/\nu_L)$ is the Reynolds number and $\mathrm{Sc}(=\nu_L/D_{TL})$ is the Schmidt number for tritium in liquid lithium.

2.2. Numerical results

Analysis was performed for typical permeation tube dimensions, which are an inner diameter of 10 mm, a wall thickness of 1.5 mm and a length of 10 m. The operating temperature of the permeation window is assumed to be 450°C. The tritium concentration in lithium is assumed to be 1 wppm, which corresponds to a tritium molar density of about $C_{A0} = 8.17 \times 10^{-2}$ mol m^{-3}.

Fig. 1 shows the relationship between the effluent concentration and tritium diffusivity of the tube materials. It is found that the effluent concentration decreases as the tritium diffusivity increases. However, when the diffusivity is larger than 1×10^{-5} m^2 s^{-1}, the effluent concentration does not change with the diffusivity, because the mass transfer coefficient limits the tritium transport in the permeation tube. This effect is not significant for the reactor permeation window, because the diffusivity in the permeation tube is used in the range 1×10^{-9} to 1×10^{-8} m^2 s^{-1}. For a diffusivity lower than 1×10^{-6} m^2 s^{-1}, the tritium transport process is dominated by the diffusion mechanism. To extract large amounts of tritium from liquid lithium, it is necessary to use a tube material that has a large diffusivity. Moreover, because the diffusivity depends on temperature, it is also necessary to operate at a higher temperature. The effluent concentration increases as the velocity increases, because the contact time between the fluid and tube wall decreases.

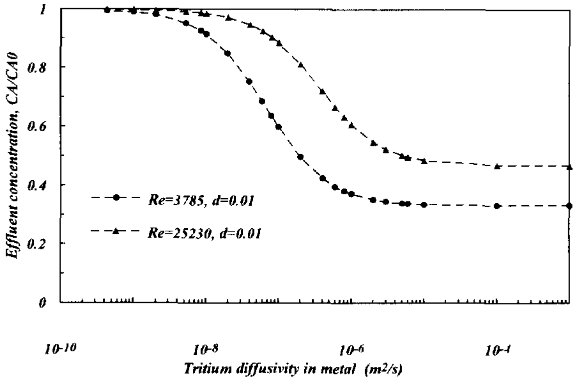

Fig. 1. Relationship between the effluent concentration and tritium diffusivity in metal.

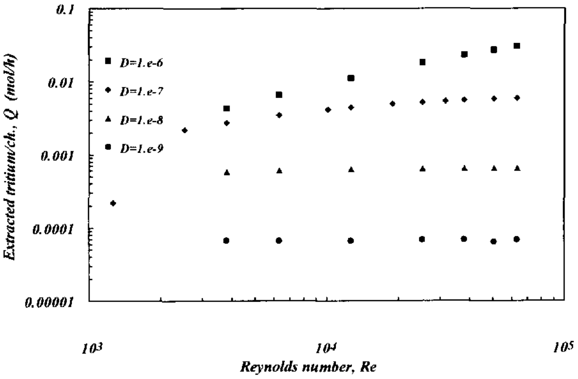

Fig. 2. Relationship between the amount of extracted tritium per channel and Reynolds number.

The amount of surface area required for tritium extraction is obtained from the numerical results. The relationship between the amount of extracted tritium, i.e. $Q(=C_{A\mathrm{out}} - C_{A\mathrm{in}})uA_f)$, and Reynolds number as a parameter of the tritium diffusivity is shown in Fig. 2. The results indicate that the amount of extracted tritium increases with increasing flow velocity for a diffusivity greater than 1×10^{-6} m^2 s^{-1}. For typical reactor permeation windows, the effect of increasing the velocity is not significant. An estimation of the surface area needed to extract 500 g per day (20.83 g h^{-1} = 3.472 mol h^{-1}) for vanadium is about 2.0×10^3 m^2 in the present case.

3. Gettering processes

3.1. Numerical model and basic equation

In the gettering processes, liquid lithium flows through the yttrium particle bed and tritium is absorbed by the yttrium particles. The continuity equation, the momentum equation of the liquid lithium, the mass conservation equation of the tritium in the particle bed, and the diffusion equation in the particle were also solved in this analysis. A typical absorbed bed is 0.25 m in diameter and 2 m in length. An yttrium particle is assumed to be spherical, with a particle diameter of 1 mm. The velocity distribution of liquid lithium in the bed is given by the steady state results of the porous flow analysis. The tritium concentration in lithium is assumed to be 1 wppm.

The governing equations based on the non-Darcian model for the velocity distribution of fully developed flow in a cylindrical packed bed are

$$\frac{\partial u}{\partial z}+\frac{1}{r}\frac{\partial (rv)}{\partial r}=0 \tag{6}$$

$$\frac{\partial u}{\partial t}+u\frac{\partial u}{\partial z}+v\frac{\partial u}{\partial r}=-\frac{1}{\rho}\frac{\partial p}{\partial z}+\frac{\partial}{\partial z}\left(\nu\frac{\partial u}{\partial z}\right)+\frac{1}{r}\frac{\partial}{\partial r}\left(r\nu\frac{\partial u}{\partial r}\right)-\frac{\nu}{K}u-C|U|u \tag{7}$$

$$\frac{\partial v}{\partial t}+u\frac{\partial v}{\partial z}+v\frac{\partial v}{\partial r}=-\frac{1}{\rho}\frac{\partial p}{\partial r}+\frac{\partial}{\partial z}\left(\nu\frac{\partial v}{\partial z}\right)+\frac{\partial}{\partial r}\left\{\frac{1}{r}\frac{\partial}{\partial r}(\nu r v)\right\}-\frac{\nu}{K}v-C|U|v \tag{8}$$

where u and v are the velocities in the axial and radial directions, $|U|$ is $(u^2+v^2)^{0.5}$, p is the pressure and ν is the kinematic viscosity of the fluid. The momentum equation takes into account friction caused by macroscopic shear (Brinkman effect) and flow inertia (Forchheimer effect). Both the permeability K and the inertia coefficient C of the porous matrix can be expressed in terms of the particle diameter and porosity from the correlations developed by Ergun [14], i.e.

$$K=\frac{d_p^2\varepsilon^3}{150(1-\varepsilon)^2} \tag{9}$$

$$C=\frac{1.75(1-\varepsilon)}{d_p\varepsilon^3} \tag{10}$$

To account for the wall effect on the packing, an exponential decrease is usually assumed to approximate the near-wall porosity variation, such as in packed-sphere beds [15,16], i.e.

$$\varepsilon=\varepsilon_0\left[1+A\exp\left\{1-\frac{N(R-r)}{d_p}\right\}\right] \tag{11}$$

where $\varepsilon_0=0.4$ is the free-stream porosity, d_p is the particle diameter, and A and N are empirical constants which depend on the packing of particles near the solid wall. The value for A is determined so that the porosity at the wall is 1.0; then $A=0.552$ is used in this calculation. The value of $N=2$ is used to represent the decay of porosity from the solid wall [16]. Fig. 3 shows the averaged porosity function calculated from Eq. (11).

The mass conservation equation of tritium is given as

$$\frac{\partial C_A}{\partial t}+\frac{\partial}{\partial z}\left(uC_A-D_{TL}\frac{\partial C_A}{\partial z}\right)+\frac{1}{r}\frac{\partial}{\partial r}\left\{r\left(vC_A-D_{TL}\frac{\partial C_A}{\partial r}\right)\right\}=-\frac{1-\varepsilon}{\varepsilon}\frac{\partial C_S}{\partial t} \tag{12}$$

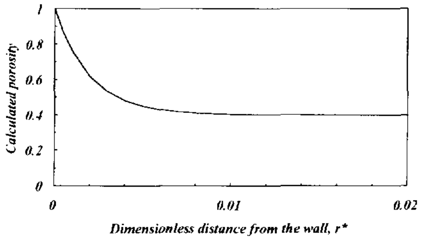

Fig. 3. Averaged porosity function (calculated from Eq. (11)).

where C_A is the tritium molar density in the liquid phase and C_S is that in the solid phase. D_{TL} is the tritium diffusivity in lithium, as given by Eq. (3).

External mass transfer through the stagnant film surrounding the particle is described by [17]

$$\frac{\partial C_S}{\partial t}=\frac{\alpha_f a}{1-\varepsilon}(C_A-C_{Ai}) \tag{13}$$

where α_f is the fluid phase mass transfer coefficient and C_{Ai} is the interfacial concentration in the fluid phase. The parameter a is defined as $a=6(1-\varepsilon)/d_p$, which gives the surface area of the packing per unit volume of the bed. For spheres, d_p is equal to the diameter.

Wilson and Geankoplis [18] investigated mass transfer between liquids and spheres in packed beds. In the ranges $0.0016<\mathrm{Re}'<55$, $165<\mathrm{Sc}<70\,600$ and $0.35<\varepsilon<0.75$, the proposed expression is

$$\varepsilon J_D=1.09\,\mathrm{Re}'^{-2/3} \tag{14}$$

In the ranges $55<\mathrm{Re}'<1500$ and $165<\mathrm{Sc}<10\,690$, they proposed the equation

$$\varepsilon J_D=0.25\,\mathrm{Re}'^{-0.31} \tag{15}$$

where J_D is the Chilton–Colburn j factor, which is $J_D=\mathrm{Sh}/(\mathrm{Re}'\mathrm{Sc}^{1/3})$. The Reynolds number is defined in terms of the diameter of the sphere (d_p), and the superficial velocity of the liquid as $\mathrm{Re}'=U'd_p/\nu$. The superficial velocity is based on the empty column cross-section and can be written in terms of the porosity and interstitial velocity as $U'=\varepsilon U$.

The balance for uptake by diffusion into the sphere is given by the non-steady state equation

$$r_p^2\frac{\partial C_S}{\partial t}=D_{TY}\frac{\partial}{\partial r_p}\left(r_p^2\frac{\partial C_S}{\partial r_p}\right) \tag{16}$$

where r_p is the radius of the particle, D_{TY} is the tritium diffusivity in yttrium, described as [11]

$$\log D'_{TY}=-2.391-2558/T\quad(\mathrm{cm^2\,s^{-1}}) \tag{17}$$

The initial and boundary conditions are

$$C_S = 0 \quad \text{at } t = 0, 0 \leqslant r_p \leqslant R \quad \text{(initial)} \tag{18}$$

$$\frac{\partial C_S}{\partial r_p} = 0 \quad \text{at } r_p = 0 \quad \text{(boundary)} \tag{19}$$

$$\begin{aligned} -D_{TY}\frac{\partial C_S}{\partial r_p} &= \alpha_f(C_A - C_{Ai}) \\ C_{Si} = \beta C_{Ai} \quad &\text{at } r_p = R \end{aligned} \tag{20}$$

The relationship between C_{Si} and C_{Ai}, i.e. the equilibrium isotherm, is obtained from assuming that the equilibrium condition is reached at the particle surface. Because there are many reported values for Sievert's constant of tritium in yttrium and lithium, we used the typical value obtained from equations in this analysis [19,20], i.e.

$$Ks_{TY} = 4.763 \times 10^{-2} \exp\left\{\frac{22900}{R_0 T}\right\} \quad (\text{wppm Pa}^{-0.5}) \tag{21}$$

$$Ks_{TL} = 3.707 \exp\left\{\frac{10104}{R_0 T}\right\} \quad (\text{wppm Pa}^{-0.5}) \tag{22}$$

At equilibrium, the tritium partial pressures in the lithium and yttrium are equal; therefore, the tritium interfacial concentration in the fluid and solid phases can be calculated using

$$\begin{aligned} C_{Si} &= \frac{Ks_{TY}}{Ks_{TL}} C_{Ai} = \beta C_{Ai} \\ &= 1.285 \times 10^{-2} \exp\left\{\frac{12796}{R_0 T}\right\} C_{Ai} \end{aligned} \tag{23}$$

In the present analysis, we introduce the following dimensionless variables:

$$z^* = z/L_{\text{ref}}$$

$$r^* = r/L_{\text{ref}}$$

$$t^* = t/(L_{\text{ref}}/U_{\text{ref}})$$

$$u^* = u/U_{\text{ref}}$$

$$v^* = v/U_{\text{ref}}$$

$$p^* = p/(\rho U_{\text{ref}}^2)$$

3.2. Numerical method

The differential transport equations are converted into a set of algebraic finite difference equations for the specified grid system using hybrid differencing schemes. A non-uniform mesh, staggered grid system is used for the packed bed to account for the wall effect. In the staggered grid system, the velocity components are calculated for the points that lie on the control volume faces; scalar quantities are calculated at the intersections of the grid points. The pressure-correction equation is solved by the SIMPLE algorithm. A detailed description for this method can be found in ref. [21]. The resulting discretization equations are solved by a line-iterative method using a tri-diagonal matrix algorithm with alternating sweep direction.

3.3. Numerical results

Fig. 4 shows the dimensionless velocity profiles at the outlet of the bed for various averaged inlet velocities. As shown, the velocity at a distance of about one-half sphere diameter from the wall is higher than that in the bulk. The peak velocity decreases as the averaged inlet velocity increases. The velocity profiles are in qualitative agreement with the variational solution obtained by Vortmeyer and Schuster [16], because the region of high velocity exists within one-half sphere diameter from the wall. When the ratio of the bed and particle diameter (d/d_p) is 250 and the particle Reynolds number $\text{Re}_p(= ud_p/\nu)$ is 2.5, their result of the calculated ratio $u_{\max}/u_{\text{center}}$ is about 9. In the case of the inlet velocity of 10 m h^{-1}, the present result of the ratio $u_{\max}/u_{\text{center}}$ is about 8.7.

Tritium breakthrough curves at various radial points in the packed bed outlet are shown in Fig. 5. The bed

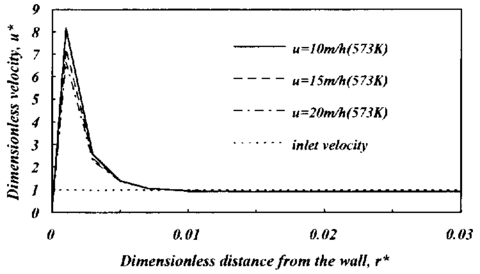

Fig. 4. Dimensionless velocity profile at the outlet of the bed.

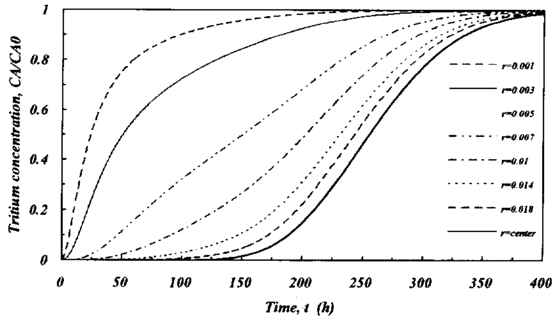

Fig. 5. Tritium breakthrough curves at various radial points: $u = 10$ m h^{-1}; $T = 573$ K.

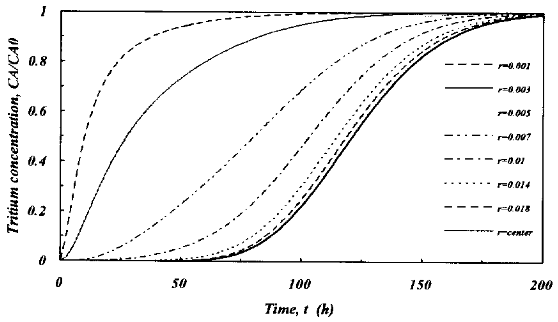

Fig. 6. Tritium breakthrough curves at various radial points: $u = 20\ \mathrm{m\ h^{-1}}$; $T = 573$ K).

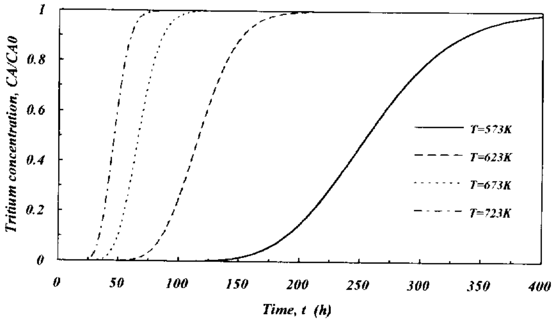

Fig. 7. Tritium breakthrough curve at the center of the bed as a function of the bed temperature.

temperature is 300°C and the averaged inlet velocity is $10\ \mathrm{m\ h^{-1}}$ ($2.78 \times 10^{-3}\ \mathrm{m\ s^{-1}}$). The time at which the bed becomes saturated with tritium in the near-wall region is shorter than that for the center of the bed. The saturation time of the bed is about 400 h at the center and about 200 h in the near-wall region in this case. Fig. 6 shows the tritium breakthrough curve when the averaged inlet velocity is $20\ \mathrm{m\ h^{-1}}$ ($5.56 \times 10^{-3}\ \mathrm{m\ s^{-1}}$). In this case, when the saturation time of the bed decreases with increasing inlet velocity, it decreases to about 200 h.

The effect of the bed temperature on the bed saturation time is shown in Fig. 7. The mass transfer coefficient increases with increasing bed temperature; at the same time, the tritium solubility in yttrium approaches that in lithium. From Eq. (23), it is found that the value of β decreases as the temperature increases. When $T = 1485$ K, $\beta = 1$, which means the interfacial concentration in the liquid phase is equal to that in the solid phase. Therefore, the tritium concentration in the particle decreases significantly as the bed temperature increases, and the saturation time of the bed also decreases.

4. Conclusions

Numerical studies were performed on tritium extraction by permeation window and gettering processes for a liquid metal breeder-blanket in a fusion reactor. In the permeation window, the effluent concentration decreases as the tritium diffusivity increases. However, when the diffusivity is larger than $1 \times 10^{-5}\ \mathrm{m^2\ s^{-1}}$, the effluent concentration does not change with diffusivity, because the mass transfer coefficient limits tritium transport in the permeation tube. This effect is not significant for the reactor permeation window. For a diffusivity lower than $1 \times 10^{-6}\ \mathrm{m^2\ s^{-1}}$, the tritium transport process is dominated by the diffusion mechanism. To extract large amounts of tritium from liquid lithium, it is necessary to use a tube material that has a large diffusivity. Moreover, as the diffusivity depends on the temperature, it is also necessary to operate at a higher temperature. The amount of extracted tritium increases with increasing flow velocity for a diffusivity greater than $1 \times 10^{-6}\ \mathrm{m^2\ s^{-1}}$. For typical reactor permeation windows, the effect of increasing the velocity is not significant. To estimate accurately the surface area of the permeation window, it is necessary to consider the effect of the resistance at the inner surface of the tube and oxidation or other surface reaction processes at the outer surface of the tube.

In the getter bed, the velocity, concentration profiles, and breakthrough curves as a function of the bed temperature and inlet velocity were obtained numerically. The near-wall region of the bed saturated with tritium in a shorter time, as a result of the higher velocity in the near-wall region. Moreover, the saturation time of the bed is highly dependent on the bed temperature. The tritium concentration in the particle decreases significantly as the bed temperature increases and the saturation time of the bed also decreases. To extract a large amount of tritium from lithium, it is necessary to operate the yttrium getter bed at low temperatures (about 300°C). Although the bed is saturated after a shorter time for higher temperature operation (400°C or more), the accumulated tritium concentration in the bed is much lower compared with the case of operating temperatures of 300°C. Therefore, it is necessary to regenerate the bed more frequently for higher temperature operations. Further analysis of regeneration process should be carried out.

Appendix A: nomenclature

a	surface area of packing per unit volume of bed ($m^2\,m^{-3}$)
A	constant in Eq. (11) (dimensionless)
A_f	flow area of tube (m^2)
C	inertia coefficient defined by Eq. (10) (m^{-1})
C_A	tritium molar density in the liquid phase ($mol\,m^{-3}$)
C_{Ai}	interfacial tritium molar density in the liquid phase ($mol\,m^{-3}$)
C_S	tritium molar density in the solid phase ($mol\,m^{-3}$)
C_{Si}	interfacial tritium molar density in the solid phase ($mol\,m^{-3}$)
d	diameter of tube or packed bed (m)
d_p	diameter of particle (m)
D_{TL}	tritium diffusivity in lithium (also D'_{TL}; $cm^2\,s^{-1}$)
D_{TY}	tritium diffusivity in yttrium (also D'_{TY}; $cm^2\,s^{-1}$)
D_{TM}	tritium diffusivity in metal ($m^2\,s^{-1}$)
J_D	Chilton–Colburn j factor (dimensionless)
K	permeability defined by Eq. (9) (m^2)
Ks_{TL}	tritium solubility in lithium (wppm $Pa^{-0.5}$)
Ks_{TY}	tritium solubility in yttrium (wppm $Pa^{-0.5}$)
L_h	length of wet periphery (m)
N	constant in Eq. (11)
p	pressure (Pa)
Q	amount of extracted tritium ($mol\,s^{-1}$)
r	radial distance in the tube or the packed bed (m)
r_p	radial distance in the particle (m)
R	radius of packed bed (m)
R_0	gas constant ($cal\,mol^{-1}\,K^{-1}$)
Re	Reynolds number (dimensionless)
Re′	Reynolds number based on the superficial velocity (dimensionless)
Re_p	particle Reynolds number (dimensionless)
Sc	Schmidt number (dimensionless)
Sh	Sherwood number (dimensionless)
t	time (s)
T	temperature (K)
u	velocity component in the axial direction ($m\,s^{-1}$)
U	interstitial velocity ($m\,s^{-1}$)
U'	superficial velocity ($m\,s^{-1}$)
v	velocity component in the radial direction ($m\,s^{-1}$)
z	axial distance in the tube or the packed bed (m)
α_f	mass transfer coefficient ($m\,s^{-1}$)
β	factor in Eq. (23)
ε	porosity (dimensionless)
ε_0	free-stream porosity (dimensionless)
ν	kinematic viscosity ($m^2\,s^{-1}$)

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