

# Development of the Tritium Transport Model for Pebbles of $\text{Li}_2\text{TiO}_3$ <sup>\*)</sup>

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Tritium transport code in  $\text{Li}_2\text{TiO}_3$  pebbles were developed in this study. In the simulation code, tritium transports in a grain, a closed-pore and an opened-pore were modeled and combined. The tritium transport in grain was modeled by the tritium diffusion, trapping/detrapping by defects, and the annihilation process of defects. The adsorption/desorption equilibrium of diffusing tritium gas molecule was modeled for the tritium transports in opened-pores. A vacant-core and shell model was used to model the tritium transport in closed-pores. The results by simulation code suggested that the tritium trapping by closed-pores would result in the shift of tritium release toward higher temperature side as dissociation of tritium gas molecule in closed-pores requires higher activation energy.

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## 1. Introduction

The design concepts of solid tritium blanket system have presented so far that the tritium breeding ratio (TBR), which is a relative value of the tritium generation rate to the tritium consumption rate, is not enough higher than unity, and it indicates that the tritium generated in blanket system must be recovered thoroughly. Many researchers suggested that the tritium release from the solid tritium breeding materials is controlled by the tritium diffusion in the grain [1–3]. Our previous study established a qualitative tritium migration model in a grain of lithium meta-titanate ( $\text{Li}_2\text{TiO}_3$ ) which is one of the promising candidate of the solid tritium breeding materials [4].

The solid tritium breeding materials will be installed in blanket systems as the form of pebbles which are the spherically sintered grains. Our previous model ignores the tritium transfer after releasing out from grains and during migrating to the pebble surface. Therefore, the tritium transfer process in pebbles should be added in the model to make the model available for an estimation of tritium inventory in actual blanket system.

The fabrication methods for pebbles of the solid tritium breeding materials have been widely investigated so far. The mechanical properties and the microstructures of fabricated pebbles were surveyed in those works to estimate the fabrication utility [5–7]. Microstructures of the cross-section of pebbles showed not only many grains combined with each other but also the voids which are so-called the pores. The pebbles were usually fabricated at

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## Nomenclature

$C$ : concentration [ $\text{m}^{-3}$ ]	<i>Subscript</i>
$N$ : the number of subject[-]	g: grain
$P$ : Partial pressure [Pa]	p: pebble
$\Omega$ : defect concentration [ $\text{m}^{-3}$ ]	OP: opened-pore
$W$ : concentration of constituent atoms [ $\text{m}^{-3}$ ]	CP: closed-pore (vacant core + shell)
$\psi$ : function	V: vacant core of closed-pore
$H$ : tritium retention [-]	m: mobile
$G$ : source term [ $\text{m}^{-3} \text{s}^{-1}$ ]	t: trapping
$\gamma_{\text{ads}}$ : adsorption capacity [ $\text{m}^{-3}$ ]	dt: detrapping
$D$ : diffusion rate [ $\text{m}^2 \text{s}^{-1}$ ]	ads: adsorbed state
$k$ : reaction rate [ $\text{s}^{-1}$ ]	gas: gas state
$S$ : solubility [ $\text{m}^{-3} \text{Pa}^{-1/2}$ ] ( $P > 1 \text{Pa}$ )	atom: atomic state
$S$ : solubility [ $\text{m}^{-3} \text{Pa}^{-1}$ ] ( $P < 1 \text{Pa}$ )	knud: Knudsen diffusion regime
$n$ : factor of tritium chemical form ( $n=1,2$ )	da: defect annihilation
$V$ : volume [ $\text{m}^3$ ]	
$M$ : molecular weight [ $\text{kg mol}^{-1}$ ]	
$\lambda$ : mean free path [m]	
$\rho_{\text{th}}$ : theoretical density of pebble [ $\text{m}^{-3}$ ]	
$\rho_{\text{act}}$ : actual density of pebble [ $\text{m}^{-3}$ ]	
$R_{\text{gas}}$ : gas constant [ $\text{J K}^{-1} \text{mol}^{-1}$ ]	
$k_{\text{B}}$ : Boltzmann constant [ $\text{J K}^{-1}$ ]	
$T$ : temperature [K]	
$R$ : radius [m]	
$d$ : diameter [m]	
$r$ : radial coordinate [m]	
$t$ : time [s]	
$\varphi$ : porosity [-]	
$\tau$ : tortuosity [-]	
$\alpha$ : defect generation constant	
$i$ : type of defect	

**\*Bold character expresses a matrix**

around 1100 K to avoid lithium evaporation [8]. Indeed, the relative density of pebbles, which is defined as the ratio of an actual pebble density to a theoretical pebble density, was usually 70 ~ 90 %.

The pores were distinguished into the opened-pores and the closed-pores. The former is pores eventually connecting to the surface of pebbles, and the latter is pores enveloped by grains. It is considered that tritium in these two types of pores will behave differently.

For the first step of model development, it was assumed that all pores existing in the  $\text{Li}_2\text{TiO}_3$  pebbles distribute uniformly in the present work. Then, the simulation code for tritium release behaviors was developed. Tritium release behaviors predicted by the simulation code were compared with those obtained in out-pile thermal desorption experiment in the previous works.

## 2. Modeling

### 2.1 Tritium transport in grain

Tritium transport in grain was modeled with McNabb-Foster model combined with the defect annihilation/generation model as follows [4].

$$\frac{\partial C_{g,m}}{\partial t} = D_g \left( \nabla_{r_g}^2 C_{g,m} + \frac{2}{r_g} \nabla_{r_g} C_{g,m} \right) - \sum_{i=1}^n \frac{\partial C_{g,t}^i}{\partial t} + G_g = \Psi_{g,1}, \quad (1)$$

$$\frac{\partial C_{g,t}^i}{\partial t} = -k_{dt}^i C_{g,t}^i + k_t^i C_{g,m} \left( \frac{\omega^i - C_{g,t}^i}{W} \right) = \Psi_{g,2}, \quad (2)$$

$$\frac{\partial \omega^i}{\partial t} = -k_{da}^i \omega^i + \alpha^i G_g = \Psi_{g,3}. \quad (3)$$

The Eq. 1 expresses the tritium diffusion in spherical grain with trapping/detrapping reactions and tritium generation by neutron irradiation. The Eq. 2 shows the linear combination of tritium detrapping rate and trapping rate in the defects. The Eq. 3 indicates the linear combination of annihilation and generation rates of defects. The tritium trapping rate, detrapping rate and the annihilation rate of defects should depend on the type of defects. It was suggested that the defects working as the tritium trapping site should be O-centers, which are the oxygen hole centers, and other types of defects weakly interact with tritium in  $\text{Li}_2\text{TiO}_3$  [9]. Therefore, tritium interaction with O-centers was only considered in the model ( $i = 1$ ).

### 2.2 Tritium transport in opened-pores

#### 2.2.1 Definition of opened-pores

Tritium migrated in grain will finally reaches to surface and desorbed out into pores with forming gas molecules. Tritium as the form of tritiated water (HTO,  $\text{T}_2\text{O}$ ) and/or molecule (HT,  $\text{T}_2$ ) in the opened-pores should diffuse with interacting on the inner surface of the opened-pores, therefore, the adsorption-desorption reactions will occur. Besides, the diffusion rate of gas molecules in opened-pores should be depended on the size of the opened-pores. Assuming the simple model where the opened-pores are formed uniformly along the all of grains in the pebble, the decrease of pebble density to the theoretical density can be expressed by the increase of grain size due to the surrounding open-pores. For this assumption, following equation can be obtained.

$$\frac{4}{3}\pi(R_g + R_{OP})^3 N_g = \frac{4}{3}\pi R_p^3. \quad (4)$$

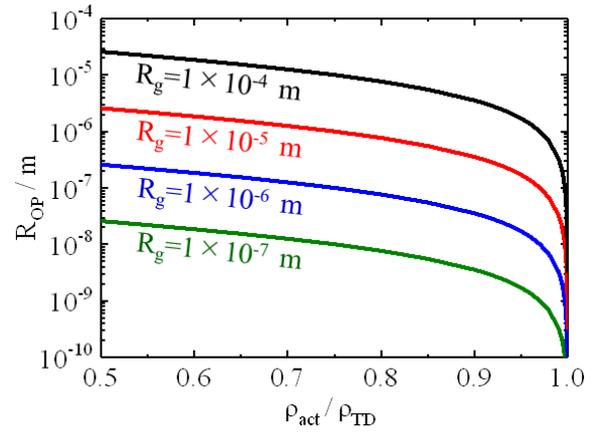


Fig. 1 Estimation of  $R_{OP}$  as a function of  $R_g$  and porosity of pebble.

Also, the theoretical density of pebble can be expressed as follows.

$$\frac{4}{3}\pi R_g^3 N_g = \frac{\rho_{act}}{\rho_{TD}} \frac{4}{3}\pi R_p^3. \quad (5)$$

The combination of Eqs. 4 and 5 gives following equation.

$$R_{OP} = R_g \left[ \left( \sqrt[3]{\frac{\rho_{act}}{\rho_{TD}}} - 1 \right)^{-1} \right]. \quad (6)$$

The porosity in pebbles is due to closed-pores as well as opened-pores. Therefore, the Eq. 6 can be converted as follows.

$$R_{OP} = R_g \left[ \left( \sqrt[3]{\frac{\rho_{act}(1 - \varphi_{CP})}{\rho_{TD}}} - 1 \right)^{-1} \right]. \quad (7)$$

Figure 1 shows the estimated radius of opened-pores with the change of the grain size in pebble. Note that the contribution of closed-pores is eliminated in Fig. 1. In other words, the pebble only with opened-pores is assumed in this figure. The radius of opened-pores decreased with the decrease of grain size. Also, the radius of opened-pores in the pebble with high density should be small.

#### 2.2.2 Diffusion mechanism

Diffusion of gas molecule in porous media has been studied so far for many research fields [10–12]. Diffusing gas molecules collide with the inner surface of opened-pores as well as other gas molecules such as helium when helium is used in blanket system as the purge gas. Diffusion mechanisms of gas molecule in porous media usually estimated with the mean free path. The mean free path in the mixture gas of two molecules (molecules 1 and 2) can be expressed as follows.

$$\lambda_1 \approx \frac{k_B T}{P_2 \pi (R_1 + R_2)^2} \left( \frac{M_2}{M_1 + M_2} \right)^{\frac{1}{2}}. \quad (8)$$

The Eq. 8 is applicable to estimate the mean free path of tritium gas molecule in pebble in the blanket system where the relatively very small amount of tritium gas molecules

(molecule 1 in Eq. 8) is mixed with relatively huge amount of helium gas (molecule 2 in Eq. 8) as the purge gas.

A typical method to distinguish the diffusion mechanism of gas molecules is utilizing the Knudsen number,  $Kn$ . The Knudsen number is defined as the ratio of the mean free path of gas molecule to the diameter of opened-pores. When the Knudsen number is larger than 10, the diffusion rate of gas molecule can be governed as the Knudsen diffusion mechanism. In the Knudsen diffusion mechanism, the diffusion of gas molecules is controlled by the consecutive collisions of gas molecules with inner surfaces of opened-pores as the mean free path of gas molecule will be much longer than the distance between the inner surfaces. Figure 2 shows the Knudsen number of tritium gas molecules as HT in the opened-pores. For the calculation, it is adopted that the radius of helium is 0.032 nm and that of HT is 0.028 nm [13]. Tritium gas molecule as HT and the partial pressure of helium in opened-pores as 0.1 MPa were assumed in Fig. 2. The assumption for the partial pressure of helium is based on the design works for solid breeder blanket where helium with the partial pressure of 0.1 - 0.2 MPa flows as purge gas [14, 15]. The Knudsen number increased as the radius of opened-pores decreased. The Knudsen number also increased with increasing temperature. As found in Fig. 2, the Knudsen number is below 10 when the radius of opened-pores is around  $1 \times 10^{-6}$  m. This radius of opened-pores is, for example, corresponding to the grain radius of  $1 \times 10^{-5}$  m with the pebble density of around 75 %, according to Fig. 1. However, it was considered in this study to simulate the transfer of tritium gas molecule in the opened-pores in pebbles composed with any radius of grains and any value of pebble density. This assumption can be justified by following reasons. At first, the grain of solid tritium breeding materials should be smaller for a quick tritium release. Also, the pebble density should be enough high for higher TBR. The temperature of blanket system is high. The partial pressure of helium in the opened-pores should be much lower than ambient pressure in blanket system due to a pressure loss into opened-pores. Finally, the existence of closed-pores is excluded in Fig. 2. For these reasons, the Knudsen number of above 10 can usually be achievable in actual blanket condition.

Then, the diffusion rate of gas molecule can be estimated as the following Knudsen diffusion equation [16].

$$D_{\text{Knud}} = \frac{2}{3} R_{\text{OP}} \sqrt{\frac{8R_{\text{gas}}T}{\pi M}}. \quad (9)$$

As found in this equation, the rate of Knudsen diffusion is dependent on the molar weight of gas molecule and the diameter of opened-pores.

### 2.2.3 Transport model in opened-pores

The tritium transport model in the opened-pores can be expressed as the Knudsen diffusion of tritium gas molecules in spherical media through opened-pores. Also,

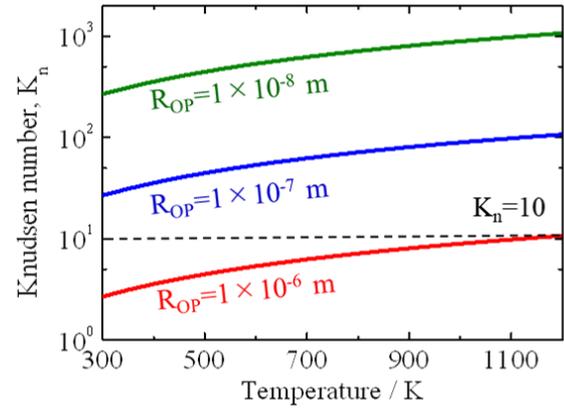


Fig. 2 Knudsen numbers of HT diffusion as function of  $R_{\text{OP}}$ .

the tritium gas molecule will experience the adsorption-desorption equilibrium reaction on the inner surface of opened-pores. The adsorption-desorption equilibrium is a linear combination of adsorption rate and desorption rate. The former depends on the adsorption capacity which is determined by the partial pressure of gas molecule and the surface area of opened-pores. Finally, the tritium transport process in opened-pores can be expressed as follows.

$$\frac{\partial C_{\text{op,m}}}{\partial t} = \frac{\varphi_{\text{op}}}{\tau} D_{\text{Knud}} \left( \nabla_{r_g}^2 C_{\text{op,m}} + \frac{2}{r_g} \nabla_{r_g} C_{\text{op,m}} \right) - \frac{\partial C_{\text{op,ads}}}{\partial t} + \frac{G_{\text{op}}}{n} = \Psi_{\text{op},1}, \quad (10)$$

$$\frac{\partial C_{\text{op,ads}}}{\partial t} = k_{\text{ads,op}} C_{\text{op,m}} \left( \frac{\gamma_{\text{op,ads}} C_{\text{op,ads}}}{\gamma_{\text{op,ads}}} \right) - k_{\text{des,op}} C_{\text{op,ads}} = \Psi_{\text{op},2}. \quad (11)$$

The tortuosity can be estimated by that opened-pores are formed along the grains discussed in the section 2.2.1, and then was assumed as  $\pi/2$ , which corresponds to the ratio of the half circumference of grain to the diameter of grain. The factor of tritium chemical form,  $n$ , in the source term of Eq. 10 is two when tritium desorbs as  $T_2$  or  $T_2O$  into opened-pores, and it will be one when tritium desorbs as HT or HTO under the presence of hydrogen isotopes. In Eq. 11, the adsorption capacity of water vapor was reported elsewhere [17], and that for hydrogen gas molecule such as HT was assumed to be negligible in this work.

### 2.3 Tritium transport in closed-pores

Tritium gas molecule in closed-pores cannot diffuse to surface of pebble as far as it dissociates and is absorbed into grains surrounding closed-pores. In this work, the spherical vacant-core and the shell models were applied for simplicity. In this model, the spherical vacant core, which meaning the closed-pores, are enveloped by larger spherical shell, which consists of  $Li_2TiO_3$  grains. The release processes of tritium gas molecule in this model are, (i) dissociative adsorption of tritium gas molecule on the inner surface of a closed-pore, (ii) absorption of tritium into grains, (iii) diffusion of tritium in grain to shell-surface,

(iv) desorption of tritium by recombination. The modeling of (i) and (ii) was carried out as that the top-most inner surface of a closed-pore is under an equilibrium between tritium gas molecule and solute tritium atoms. The kinetics of the tritium desorption process on the outer surface was excluded from the model as this process would be much faster than other processes according to our previous study [4]. Then, the following equations were obtained.

$$\begin{aligned} \frac{\partial N_{\text{cp,gas}}}{\partial t} \Big|_{r \leq R_V} &= -\frac{1}{\partial t} V_V \left( S \sqrt{\frac{N_{\text{cp,gas}}}{nV_V} R_{\text{gas}} T} \right) \\ &+ \frac{1}{\partial t} V_V C_{\text{cp,atom}} \Big|_{r=R_V} + \frac{C_{\text{cp}}}{n} - \frac{\partial H}{\partial t} \\ -4\pi r_{\text{cp}}^2 D_g \nabla_{r_g} C_{\text{cp,atom}} \Big|_{r=r_{\text{cp}}} &= \Psi_{\text{cp},1} \Big|_{r \leq R_V}, \end{aligned} \quad (12)$$

$$N_{\text{cp,gas}} \Big|_{R_V < r \leq R_{\text{cp}}} = 0 = \Psi_{\text{cp},1} \Big|_{R_V < r \leq R_{\text{cp}}}, \quad (13)$$

$$C_{\text{cp,atom}} \Big|_{r < R} = 0 = \Psi_{\text{cp},2} \Big|_{r < R_{\text{cp}}}, \quad (14)$$

$$\begin{aligned} \frac{\partial C_{\text{cp,atom}}}{\partial t} \Big|_{r=R_V} &= \frac{1}{\partial t} \left( S \sqrt{\frac{N_{\text{cp,gas}}}{nV_V} R_{\text{gas}} T} \right) \\ - \frac{1}{\partial t} C_{\text{cp,atom}} \Big|_{r=R_V} &= \Psi_{\text{cp},2} \Big|_{r=R_V}, \end{aligned} \quad (15)$$

$$\begin{aligned} \frac{\partial C_{\text{cp,atom}}}{\partial t} \Big|_{R_V < r \leq R_{\text{cp}}} &= D_g \nabla_{r_{\text{cp}}}^2 C_{\text{cp,atom}} \\ + D_g \frac{2}{r_{\text{cp}}} \nabla_{r_{\text{cp}}} C_{\text{cp,atom}} &= \Psi_{\text{cp},2} \Big|_{R_V < r \leq R_{\text{cp}}}. \end{aligned} \quad (16)$$

The first two terms in the right side of Eq. 12 express the balance between tritium gas molecule and absorbed tritium on the top-most inner surface of a closed-pore. The fourth and fifth terms of Eq. 12 indicate the rate of tritium retention in the shell and the tritium release rate from the shell. The Eq. 13 means tritium gas does not exist in the shell. In contrast, the Eq. 14 indicates tritium atom does not exist in vacant core. Eq. 16 indicates tritium atom diffuses in shell to the surface of a closed-pore.

### 3. Simulation Code

The simulation code was constructed by combining the kinetic models of tritium transports in a grain, an opened-pore and a closed-pore. In the simulation code, it was assumed that tritium should be generated in a grain uniformly as the neutron flux in grain would be uniform. This assumption leads the uniform distribution of defects in a grain. Tritium is generated in grains, and migrates to pores. Tritium hardly transports from a closed-pore to another closed-pore as the probability that closed-pores neighbor each other should be negligible. Therefore, tritium in grain will transport to opened-pores or closed-pores, and tritium in closed-pores transports to opened-pores only. Tritium in opened-pores will diffuse to the surface of pebble and release out consequently. The source terms as tritium generation rate for grain depends on the

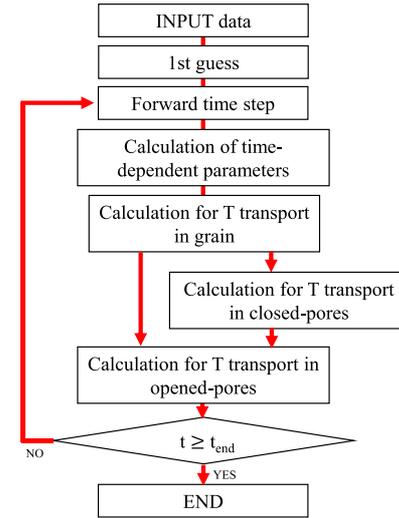


Fig. 3 Flowchart of tritium transport code in this work.

neutron fluence, and those for opened-pores and closed-pores depend on the amount of grains surrounding these pores. Therefore, the source terms for these pores were expressed as the products of the tritium release rate from a grain and the amount of grains around these pores. The source terms for opened-pores were also added by the tritium release rate from closed-pores neighboring opened-pores.

The flowchart of the simulation code prepared in this work is shown in Fig. 3. For the calculation of tritium transports in grains, closed-pores and opened-pores, the implicit finite differentiation method was used. In this method,  $x_j - \Delta t \Psi_j - x_{j-1} = 0$  is always satisfied ( $x$  is variables and  $j$  is time step) at any position for all simultaneous equations. The converged results of simultaneous equation were obtained by Newton-Raphson methods combined with Gauss-Jordan elimination calculation.

In the calculation, the kinetic parameters such as tritium diffusion rate, detrapping/trapping rates and defect annihilation rate were used as reported elsewhere [4]. The presence of hydrogen isotopes was assumed as hydrogen isotopes will be used as a purge gas in the blanket for fast tritium recovery. Therefore, most of tritium will release as gas molecular form (HT) as reported in the in-pile tritium recovery experiment [18]. The solubility of tritium gas molecule in  $\text{Li}_2\text{TiO}_3$  was roughly reported in ref. [19]. It was assumed that the tritium solubility depends on Henry's law when the partial pressure of tritium gas molecule in closed-pore is less than 1 Pa where the Sievelts's law is no longer applicable in such low pressure.

### 4. Results and Discussion

Figure 4 shows the tritium release behaviors from  $\text{Li}_2\text{TiO}_3$  pebbles estimated by the simulation code with different relative density. In this simulation, the radius of grain, pebble and closed-pores were assumed as  $1.5 \times 10^{-6}$ ,  $1.0 \times 10^{-3}$  and  $5.0 \times 10^{-6}$  m, respectively. The ki-

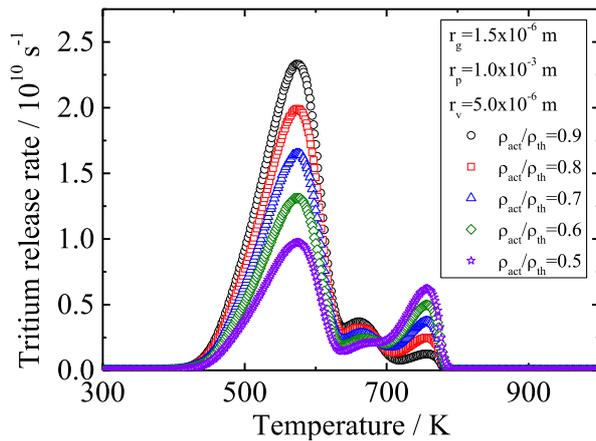


Fig. 4 Tritium release behaviors estimated by the simulation code assuming various pebble densities. Neutron fluence:  $3.3 \times 10^{15} \text{ cm}^{-2}$ , Heating rate: 5 K/min.

netics parameters for tritium migration in grain was used as reported in previous work [4]. Among these parameters, the pre-exponential factor of tritium trapping reaction was only a free-parameter, and other parameters were evaluated by the experiments. This pre-exponential factor of tritium trapping reaction was optimized by the fitting of tritium release spectrum from the  $\text{Li}_2\text{TiO}_3$  powders with the model consisting of Eqs. 1 - 3 [4]. The kinetic parameters of tritium migration in opened-pores and closed-pores were as explained in the sections 2.2 and 2.3, respectively. With decreasing the relative density of a pebble, the tritium release rate in lower temperature region was decreased and tritium release peak at 750 K was increased. In our previous study carried out for thermal neutron irradiated  $\text{Li}_2\text{TiO}_3$  grain with the grain radius of  $1.5 \times 10^{-6}$  showed a major tritium release peak at around 580 K [4]. The tritium release peak at around 580 K was assigned as the tritium release governed by the tritium diffusion process in  $\text{Li}_2\text{TiO}_3$  structure. As found in Fig. 4, a major tritium release peak was observed around 580 K. This indicates that the tritium diffusion process in  $\text{Li}_2\text{TiO}_3$  grain is a dominant process for tritium release from a pebble. Tritium release peak around 680 K was assigned as tritium trapped as hydroxyl groups [9]. Tritium release peak around 750 K should be caused by the tritium trapping in pores because this peak was not observed in the experiments carried out for  $\text{Li}_2\text{TiO}_3$  powder sample. In particular, the closed-pore should work as strong trapping site for tritium in pebbles as tritium gas in closed-pore needs a dissociation, which usually requires higher energy. These trends that a tritium release peak for pebbles shifts toward higher temperature side although a large part of tritium release is still controlled by the tritium diffusion process in grains, are almost consistent with the experimental data for  $\text{Li}_2\text{TiO}_3$  as well as  $\text{Li}_4\text{SiO}_4$  pebbles conducted by Kinjo and Munakata [20, 21]. Their experiments for  $\text{Li}_2\text{TiO}_3$  pebble showed that tritium mainly releases in lower temperature around 600 K, and the tritium release rate gradually decreased up to 900 K. As compared

to the results by a simulation code as found in Fig. 4, the tritium release continuing to higher temperature observed in the experiments would be caused by the presence of closed-pores in pebble. The size and distribution of pores in actual pebble should not be uniform, therefore, the tritium release would not appear as a peak as found in Fig. 4.

The simulation code prepared in this work could evaluate the tritium trapping effects in closed-pores. On the other hand, the same size and the uniform distribution of pores assumed in the simulation code should produce a sharp tritium release peak that is inconsistent with the experimental data. Also, the simulation code here only estimates the tritium release behavior from one pebble. In the actual blanket system, a number of pebbles will be installed. It can be considered that tritium released from a pebble will interact with other pebbles when they transport through blanket system, which process was not taken into account here. Further developments of tritium transport code are required to make the code applicable to the actual fusion reactor.

## 5. Conclusion

Tritium transport code for  $\text{Li}_2\text{TiO}_3$  pebbles were developed by modeling tritium transport processes in grain, closed-pores and opened-pores. The tritium transport in grain was modeled by the tritium diffusion, trapping/detrapping by defects, and the annihilation process of defects. The adsorption/desorption equilibrium of diffusing tritium gas molecule was modeled for the tritium transports in opened-pores. A vacant-core and shell model was used to model the tritium transport in closed-pores. The results by simulation code suggested that the tritium trapping by closed-pores would result in the shift of tritium release temperature toward higher temperature side.

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