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Influence of hydrogen addition to a sweep gas on tritium behavior in a blanket module containing Li₂TiO₃ pebbles

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HIGHLIGHTS

• Mass balance equations of H₂, H₂O, T₂ and T₂O in a Li₂TiO₃ pebble bed were numerically calculated.

- In the temperature rising process, the pebbles were exposed to water vapor of relatively high concentration.
- Tritium permeation rate to cooling water reduced with increasing hydrogen concentration in the sweep gas.
- Tritium inventory in the grain bulk and the grain surface occupied 99.6% of total inventory.

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1. Introduction

From viewpoints of the design of the blanket system and the safety control of the bred tritium, the understanding of tritium behavior in the solid blanket module is an important issue. The bred tritium diffuses in the grain of Li ceramics and is released to the grain boundary via surface reactions on the grain. Nishikawa et al. reported that the chemical form of the release tritium is decided by the competitive reaction of water desorption and isotope exchange reactions with H_2 and H_2O . The numerical calculation based on Nishikawa's model can reproduce well the tritium release curve from solid breeders in out-of-pile experiments [1]. Additionally, Kinjyo et al. estimated tritium behavior in the operational condi-

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ABSTRACT

Hydrogen addition to a sweep gas of a solid breeder blanket module has been proposed to enhance tritium recovery from the surface of the breeders. However, the influence of hydrogen addition on the bred tritium behavior is not understood completely. Tritium behavior in the simplified blanket module of Li₂TiO₃ pebbles was numerically calculated considering diffusion in the grain bulk, surface reactions on the grain surface and permeation through the cooling pipe. Although a partial pressure of T₂ increases with increasing a partial pressure of H₂ in the sweep gas, it was estimated that tritium permeation rate to the cooling water decreases. Additionally, the release duration of water vapor generated by the reaction of the pebbles and hydrogen is shortened with increasing a partial pressure of H₂. Tritium inventory in the grain bulk and the grain surface occupies 99.6 % of total tritium inventory in the blanket module. © 2016 Elsevier B.V. All rights reserved.

tion of ITER-TMB assuming the simplified blanket module without cooling pipes [1].

From viewpoints of fusion safety and fuel self-sufficiency, the estimation of the bred tritium permeation into the coolant through the cooling pipes is an important issue. The chemical form of the released tritium influences strongly on the permeation of the bred tritium to the coolant through the cooling pipes because the permeation rate of tritiated water vapor is much smaller than that of molecular tritium [2]. The tritium recovery as water vapor may be preferable from a viewpoint of suppression of tritiuated water was concentrated and converted to molecular form, becomes large. Additionally, the presence of water vapor may promote Li mass loss [3] and oxidize structural materials. Tritium recovery as molecular form is considered to be valid choice comprehensively.

In this work, tritium behavior in the simplified solid blanket module including permeation into the coolant were numerically

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Fig. 1. Illustration of calculation model.

calculated based on Nishikawa's model by using mass transfer parameters reported in literatures for Li_2TiO_3 . The impacts of hydrogen concentration in the sweep gas on tritium behavior were discussed.

2. Calculation model

2.1. Calculation region in the blanket module

The design of blanket modules for DEMO and commercial fusion reactor is under consideration at this time. Therefore the simplified blanket module are assumed here by referring [4]. The illustration of the module is schematically shown in Fig. 1. The size of the module is 200 cm in width and 60 cm in height. The calculation region is 5 cm in length between cooling pipes. The pebbles of Li₂TiO₃ are packed in the module with 0.65 in a packing ratio. The heat transfer is not calculated in this work. The maximum temperatures in the packed bed is 900 °C at the center and is 300 °C at the both edge. The following function is assumed to input the temperature distribution.

$$T_{\max}(x) = 235 + 532x - 1.06 \times 10^6 x^2, \tag{1}$$

where T_{max} is a maximum temperature [°C]at the position, *x* is a distance from one side of the packed bed. The temperature rises from 20 °C to the maximum temperature with a ramping rate of 1 °C/s. Each partial pressure of four components in the sweep gas is assumed to be uniform in the *x* direction shown in Fig. 1.

2.2. Cooling pipes

The structural material of cooling pipes is F82H. The total number of pipes are 160, which are inserted in the both edge of the packed bed. The inner diameter of a pipe is 8 mm, the thickness is 1 mm and the length is 60 cm. The flow rate of cooling water is 3 m/s. The temperature of pipes are uniformly same as the edge temperature of the packed bed. Therefore the temperature rises from 20 °C to 300 °C with time.

2.3. Tritium behavior in Li₂TiO₃ pebble bed

When tritium co-exists with H₂ and H₂O, tritium exists mainly as HT and HTO practically. However, for simplicity, four components for T₂, T₂O, H₂ and H₂O were calculated. The concentration of HT or HTO can be estimated by $2 \times T_2$ concentration or $2 \times T_2$ O concentration when H₂ and H₂O concentrations are sufficiently larger than T₂ and T₂O concentration. In the calculation of tritium release from the grain bulk to the sweep gas, the following mass transfer steps were taken into account.

Table 1

Mass transfer parameters. R is the gas constant [J/mol/K]. A_{BET} is the BET surface area $[m^2/g]$. P is the partial pressure of water vapor [Pa], T is the temperature [K].

Diffusion coefficient: Li2TiO3 [5] $D = 5.0 \times 10^{-3} \exp(-160000/RT) [m^2/s]$ Rate constant for tritium transfer at inter facial layer: Li₂TiO₃ [5] $k_{\rm ifl} = 1.0 \times 10^{-10} \exp\left(-15000/RT\right) [{\rm m/s}]$ Capacity of physically adsorbed water: Li₂TiO₃ [6] $Q_{ad,p} = 1.5 \times 10^{-7} A_{BET} P^{1/2} \exp(134000/RT)$ [mol/mol] Capacity of chemically adsorbed water: Li2TiO3 [6] $Q_{\rm ad,c} = \frac{3.7 \times 10^{-21} A_{BET} \exp(174600/RT)}{1.0+1.1 \times 10^{-16} \exp(174600/RT)} \text{ [mol/mol]}$ Adsorption/desorption rate constant: LiAlO₂ [7] $k_{\rm ad} = 1.02 \exp\left(-14650/RT\right) \,[{\rm m/s}]$ $k_{\rm des} = 1.15 \exp\left(-12600/RT\right) [m/s]$ Mass transfer coefficient for EX 1: Li₂TiO₃ [8] $k_{\text{ex1}} = 1.67 \times 10^{-2} \exp(-115500/RT) [\text{m/s}]$ Mass transfer coefficient for EX 2: Li₂ZrO₃ [9] $k_{\rm ex2} = 8.5 \times 10^{-6} \, [{\rm m/s}]$ Water formation capacity: Li₂TiO₃ [10] $1.4 \times 10^8 \exp\left(-168000/RT\right)$ $1.0 + 1.0 \times 10^{11} \exp(-168000/RT)$ [mol/mol] $2.5 \times 10^2 \exp(-130000/RT)$ $+2.0 \times 10^{-4}$ $1.0 + 1.3 \exp(-130000/RT)$ Water formation reaction rate constant: Li₂TiO₃ [10] $k_{\rm wf} = 3.0 \times 10^{-3} \exp\left(-4000/RT\right) [m^3/mol/s]$

- 1. Diffusion of bred tritium in the grains.
- 2. Mass transfer from the grain bulk to the grain surface.
- 3. Desorption and adsorption of water vapor on the grain surface.
- 4. Isotope exchange reaction between $H_2(T_2)$ in the sweep gas and $T_2O(H_2O)$ on the grain surface (Isotope exchange reaction 1:EX1)
- Isotope exchange reaction between H₂O (T₂O) in the sweep and T₂O (H₂O) on the grain surface. (Isotope exchange reaction 2:EX2)
- 6. Water formation reaction on the grain surface.

The mass balance equations are summarized in [5]. In Nishikawa's tritium release model, the interfacial layer is placed between the grain bulk and the grain surface as a mass transfer resistance. The bred tritium diffuses in the grain bulk and goes into the interfacial layer and then transfers to the surface. The bred tritium released to the surface exchanges with hydrogen existing as adsorbed water or structural water. After that, tritium on the surface is released via surface reactions. The mass transfer in the network of pores in the pebbles and the mass transfer in fluid film on the pebble surface are ignored. Nishikwa's model does not suppose the case that the amount of tritium released from the interfacial layer to the surface is larger than the amount of hydrogen on the surface. In this calculation, it is assumed that the amount of tritium beyond the amount of hydrogen on the surface is released as T₂. When tritium is generated by the nuclear reaction of Li and neutron, the oxygen atom combined with Li is freed. When the oxygen atom diffuses to the surface, the generated tritium may react with oxygen on the surface and be released as T_2O , or the oxygen atom may combine with other Li or Ti and remain in the grain. Experimental evidences are insufficient for this point. The quantification of the reaction rate of H_2 and $1/2 O_2$ on the solid breeder surface is necessary.

Mass transfer parameters used in this calculation are summarized in Table 1. Unfortunately, all parameters were not completed for one sample of Li_2TiO_3 . Adsorption and desorption rate constants

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