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Estimation of tritium release behavior from solid breeder materials under the condition of ITER test blanket module

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Abstract

A tritium release model from solid breeder materials has been developed. Tritium release curves estimated using this model gave good agreement with tritium release curves from various solid breeder materials obtained by out-pile experiments under various purge gas conditions in previous study. In this study, tritium release behavior from Li_2TiO_3 and $LiAlO_2$ are estimated for the neutron irradiation condition and operation sequence of the ITER test blanket module test program (neutron wall loading of 0.78 MW/m^2 and repetition of burn time for 400 s with a dwell time of 1400 s). The results are evaluated to discuss the tritium release and inventory performance of different breeder materials.
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1. Introduction

It has been pointed out that in addition to diffusion of tritium in bulk of grain, tritium transfer at surface layer and surface reactions on grain surface give profoundly effect to the tritium release behavior. To analyze the effects of the surface reactions on tritium release behavior from solid breeder materials, tritium release experiments were carried out using a purge gas with different compositions (dry N_2, N_2 with H_2, N_2 with H_2O) while applying the out-pile temperature programmed desorption techniques at the Japan Atomic Energy Research Institute (JAERI) [1–4].

2. Theory, tritium release model

It is considered that the following mass transfer processes contribute to the release of tritium bred in blanket materials [5–7]:

(1) tritium formation reaction in the crystal grain;
(2) diffusion of tritium in the crystal grain;
(3) interaction of tritium with irradiation defects in the crystal grain;
(4) resistance for tritium transfer from inner bulk layer to surface water;
(5) absorption of tritium into the bulk of the crystal grain;
(6) adsorption and desorption of tritium on the grain surface;
(7) isotope exchange reactions between gaseous hydrogen (H_2) in the gas stream and tritium on grain surfaces (isotope exchange reaction 1)

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H_2 (gas phase) $+T_2O$ (surface) $\leftrightarrow T_2$ (gas phase) $+ H_2O$ (surface);
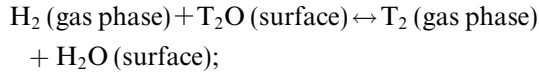
(8) isotope exchange reactions between water vapor (H_2O) in the gas stream and tritium on the grain surfaces (isotope exchange reaction 2)

H_2O (gas phase) $+T_2O$ (surface) $\leftrightarrow T_2O$ (gas phase) $+ H_2O$ (surface);
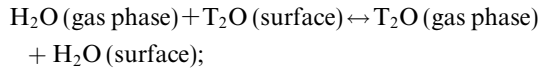
(9) water formation reactions in addition of H_2 to the blanket purge gas;

(10) transfer of hydrogen isotopes and water through pores of sintered pellets;

(11) transfer of hydrogen isotopes and water through the boundary layer formed on the surface of sintered pellets to the gas stream.

The tritium release model developed by the present authors assumes that the bred tritium in grain interior arrives at the surface layer, then tritium at surface layer is transferred to the surface water which consist of physical adsorbed water, chemical adsorbed water and structural water. Tritium on grain surface is liberated to the purge gas from surface water through such surface reactions as desorption, isotope exchange reaction between hydrogen in purge gas and isotope exchange reaction between water vapor in purge gas.

Process (4) is a newly introduced mass transfer resistance in this paper. It is assumed in this study that tritium is introduced to the interfacial layer at the rate of N_{T0} (mol/m^2 s) by diffusion and is transferred to the surface water at the rate of N_T (mol/m^2 s). The mass balance in the surface layer is expressed by the following equations:

$$A\Delta x \frac{\partial C_T}{\partial t} = AN_{T0} - a_v V N_T, \tag{1}$$

$$N_T = k\left(\frac{T}{T+H} - \frac{T_2O}{T_2O + H_2O}\right)C_T, \tag{2}$$

where C_T (mol/m^3) is the tritium concentration in the interfacial zone, a_v (m^2/m^3) is the specific surface area of the packed bed, A (m^2) is theoretical surface area of packed bed assuming that each grains are spherical and not sintered, Δx (m) is thickness of interfacial layer, V (m^3) is volume of interfacial zone in the sample bed, $T/(T + H)$ is isotopic ratio in the grain, $T_2O/(T_2O + H_2O)$ is the isotopic ratio in the surface water and k (m/s) is a rate constant.

In this study, the overall mass transfer resistance at the surface is separated into: (a) a resistance accounting for tritium transfer from the surface bulk layer of breeding material to the surface water and (b) a resistance accounting for the surface reactions related to tritium in surface water. Because of the introduction of a new mass transfer resistance at the surface layer in the new model, the same reaction rates are used for the surface reactions as those reported in previous papers [8–11]. The mass balance equations for bred tritium together with hydrogen and water vapor at the various mass transfer steps have been explained in detail in the reference [4].

The tritium diffusion in the bulk of grain is represented by an effective tritium diffusivity comprising process (2) and process (3) because irradiation defects may affect tritium migration.

3. Blanket model

The blanket model in this study is shown in Fig. 1. This model is simplified test blanket module for ITER suggested by Japan Atomic Energy Research Institute. The cross sectional area is assumed to be circle and the plug flow type mixing is assumed for purge gas. The purge gas flow rate is assumed as 54.6 L/min to give 0.5 Pa for tritium (form of HT or HTO) partial pressure in purge gas at outlet.

The tritium generation rate of 17.0×10^{-6} mol/m^3/s, and the temperature change with neutron irradiation are shown in Fig. 2, which were estimated by Japan Atomic Energy Research Institute.

The grain diameter, theoretical density of grain, void fraction of breeder first layer or amount of packed breeder material for Li_2TiO_3 are 1 μm, 0.342×10^7 g/m^3, 0.51 or 19.9 kg, respectively. And data for $LiAlO_2$ are 1 μm, 0.26×10^7 g/m^3, 0.56 or 13.6 kg.

In this paper, simulation results are presented at the point 2 mm from the purge gas inlet.

4. Results and discussion

Fig. 3 shows the tritium release to gas phase and tritium inventory for Li_2TiO_3 during the first and second neutron irradiation shot. The inventory of tritium trapped in bulk represented by curve (1) increases rapidly after neutron irradiation begins, and then it decreases since tritium diffusion from the bulk interior to interfacial layer is promoted
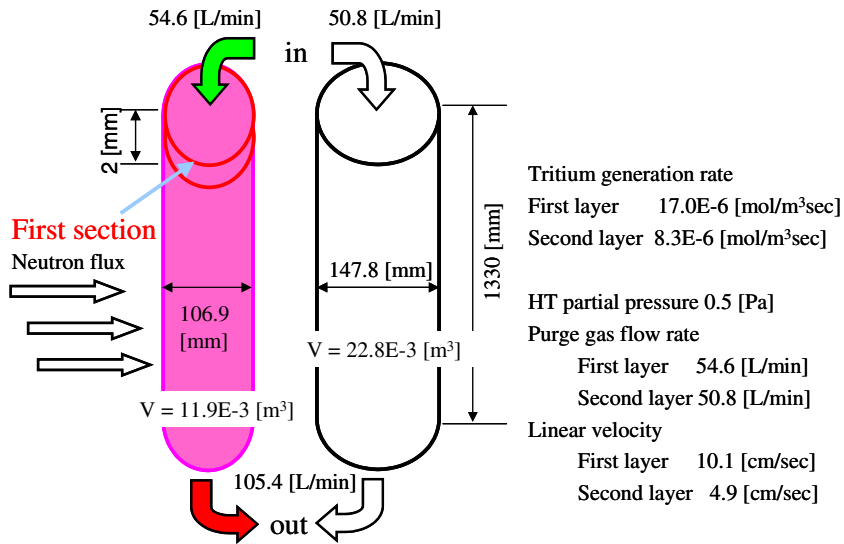
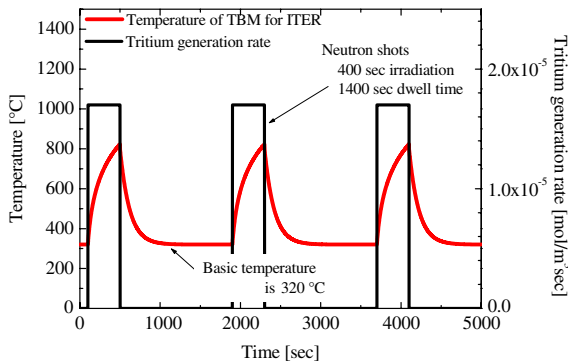Fig. 1. Blanket model for simulation of tritium release.

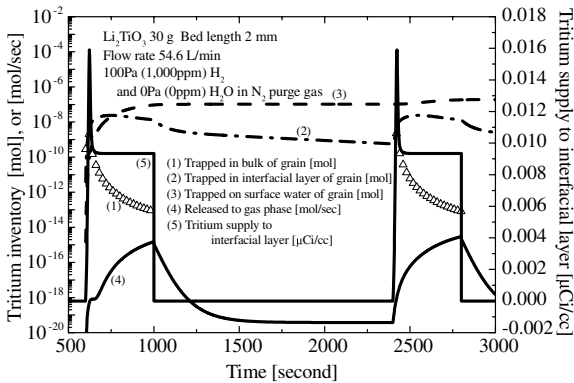Fig. 2. Temperature change and tritium generation profiles.

Fig. 3. Tritium release and inventory estimated for Li_2TiO_3 (100 Pa H_2 purge gas).

with rapid temperature rise. After the neutron irradiation stops, the inventory in bulk decreases rapidly according to transfer of tritium from bulk to interfacial layer.

The inventory in surface water represented by curve (3) increases when the neutron irradiation starts, and then it decreases only a little after neutron irradiation stops. In this calculation, it is assumed that there is no water vapor in the purge gas, therefore tritium trapped in surface water is released through only the isotope exchange reaction between tritium in surface water and hydrogen in purge gas, referred to as isotope exchange reaction 1. However it was determined that isotope exchange reaction 1 does not affect tritium release at temperatures lower than 650 K. Then tritium in surface water is not released to the gas phase under the low temperature condition after neutron irradiation stops in this purge gas composition.

The inventory trapped in interfacial layer represented by curve (2) rapidly increases according to the increase of inventory in bulk, curve (1), and then it decreases slowly until the end of neutron irradiation. After neutron irradiation, inventory in interfacial layer slowly decreases in spite of a quick decrease of inventory in the bulk of grain. It is assumed that tritium in the interfacial layer is transferred to surface water in proportion to the difference of isotopic ratio, $T/(T + H)$, in interfacial zone and, $T_2O/(T_2O + H_2O)$, in surface water as shown by Eq. (2). It is considered that the value of $T/(T + H)$ is equal to 1 in the bulk of materials. This gives no detectable absorption capacity of water or molecular hydrogen. The value of $T_2O/(T_2O + H_2O)$ is consi-

dered to be almost constant according to curve (3), therefore inventory in interfacial layer slowly decreases against decreasing of inventory in the bulk of grain after the neutron irradiation stops.

Fig. 4 shows the tritium release curve to gas phase and tritium inventory for Li_2TiO_3 during some neutron irradiation shots. Tritium inventory in surface water increases slowly during repetition of neutron irradiation for more than nine shots. Therefore it appears that 20 000 s are required to reach the steady release in this neutron irradiation condition.

The largest tritium inventory for Li_2TiO_3 is in surface water under the perfect dry purge gas condition. To decrease of the tritium inventory, it is recommended by present authors to add some amount of water vapor to the purge gas.

Fig. 5 shows the tritium release curve and tritium inventory for Li_2TiO_3 under the humid purge gas condition such as 100 Pa hydrogen and 1 Pa water vapor in purge gas. Under this purge gas condition,
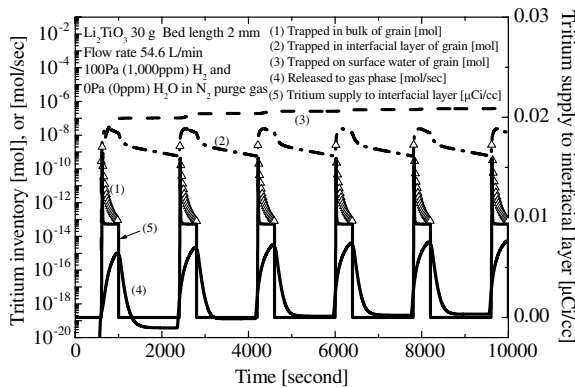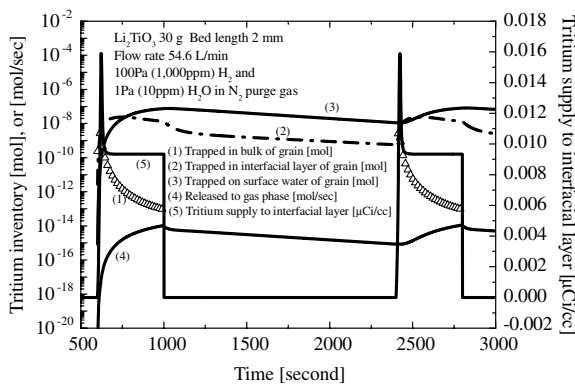
it is considered that tritium in surface water on grain is released through surface reactions such as water desorption, isotope exchange reaction 1, or isotope exchange reaction between tritium in surface water and water vapor in gas phase, referred to as isotope exchange reaction 2. The amount of tritium release represented by curve (4) is larger than that shown in Fig. 4 due to the contribution of isotope exchange reaction 2 and water desorption. Tritium in surface water is mainly released to the gas phase by isotope exchange reaction 2 under low temperature conditions after neutron irradiation stops.

It is considered that some amount of water vapor must be mixed in the actual purge gas and some amount of water is generated through the water generation reaction when hydrogen mixed gas is used as the blanket purge gas.

Fig. 6 shows the tritium release curve and tritium inventory for Li_2TiO_3 under the humid purge gas condition. The tritium inventory trapped on surface water is released to gas phase through isotope exchange reaction 2 even when the temperature is dropped after neutron irradiation stops.

When dry gas with hydrogen is used for the purge gas, tritium trapped in water on grain surface is released to gas phase through only isotope exchange reaction 1, but the reaction rate of isotope exchange reaction 1 is so slow under 673 K that the isotope exchange reaction does not give on effective contribution. It is recommended to raise the TBM temperature above 673 K to decrease the tritium inventory in the surface water if possible. It will be effective for decreasing the tritium inventory on grain surface to add some amount of water vapor to purge gas because the reaction rate of isotope exchange reaction is fast enough at the low temperature.

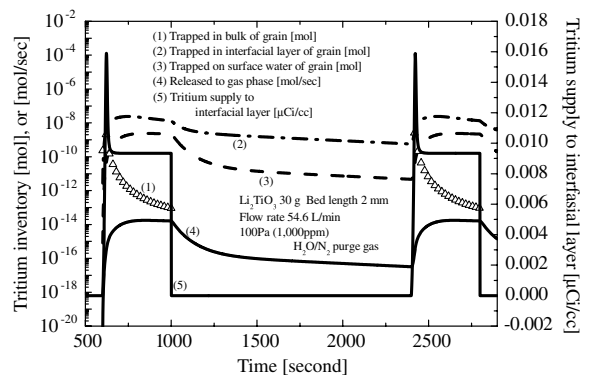Fig. 4. Tritium release and inventory estimated for Li_2TiO_3 (100 Pa H_2 purge gas).

Fig. 5. Tritium release and inventory estimated for Li_2TiO_3 (100 Pa H_2, 1 Pa H_2O purge gas).

Fig. 6. Tritium release and inventory estimated for Li_2TiO_3 (100 Pa H_2O purge gas).
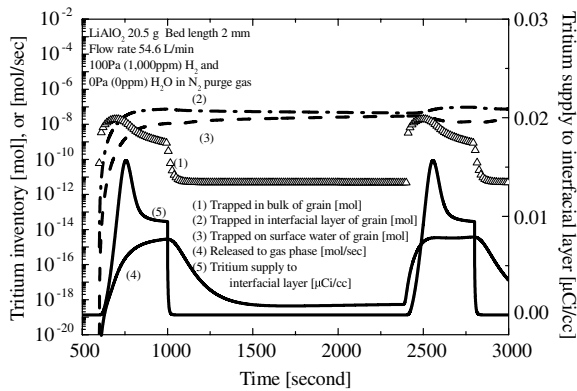
Fig. 7. Tritium release and inventory estimated for LiAlO$_2$ (100 Pa H$_2$ purge gas).

Fig. 7 shows the tritium release curve and inventory for LiAlO$_2$ purged by dry N$_2$ gas with 100 Pa hydrogen. The tritium diffusivity in the bulk of grain for LiAlO$_2$ is much smaller than any other solid breeder materials. Accordingly, tritium inventory in bulk for LiAlO$_2$ shown by curve (1) in Fig. 7 is the largest among four breeder materials examined in this study. However repetition of five shots is enough to obtain the steady state release in the case of LiAlO$_2$ because the rate of isotope exchange reaction 1 for LiAlO$_2$ is faster than that for Li$_2$TiO$_3$ in hydrogen atmosphere [12].

5. Conclusion

Tritium release behavior from solid breeder materials under the condition of ITER–TBM is estimated using a tritium release model developed for fitting with out-pile tritium release experiment. The tritium diffusivity might not give large effect to tritium inventory under ITER–TBM condition because tritium inventory in bulk of grain is much smaller than that in interfacial layer or surface water.

Using a humid purge gas is effective to decrease tritium inventory because isotope exchange reaction with water vapor is much faster than isotope exchange reaction with hydrogen, especially during dwell time between neutron shots.

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