



In-pile test of Li_2TiO_3 pebble bed with neutron pulse operation

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Abstract

Lithium titanate (Li_2TiO_3) is one of the candidate materials as tritium breeder in the breeding blanket of fusion reactors, and it is necessary to show the tritium release behavior of Li_2TiO_3 pebble beds. Therefore, a blanket in-pile mockup was developed and in situ tritium release experiments with the Li_2TiO_3 pebble bed were carried out in the Japan Materials Testing Reactor. In this study, the relationship between tritium release behavior from Li_2TiO_3 pebble beds and effects of various parameters were evaluated. The (R/G) ratio of tritium release (R) and tritium generation (G) was saturated when the temperature at the outside edge of the Li_2TiO_3 pebble bed became 300 °C. The tritium release amount increased cycle by cycle and saturated after about 20 pulse operations.

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1. Introduction

The major advantage of in situ irradiation experiments is the possibility to irradiate with neutrons in order to prove the application of a fusion reactor. The objective is to evaluate the tritium production and heat generation profiles in the tritium breeder region for the design of a fusion blanket. Recent in situ irradiation tests are CRITIC-III (Canada) [1], BEATRIX-II (US/Japan/Canada) [2–4], EXOTIC (Netherlands/EU) [5–7]. However, the experiments were evaluated from the tritium release of tritium breeders and did not simulate the structure and the operation of a fusion blanket. Engineering data on the neutron irradiation performance are indispensable to design the fusion blanket, and the knowledge of the in situ irradiation behavior of tritium breeders is limited at present.

In this study, an in-pile mockup with a Li_2TiO_3 pebble bed was developed and in situ tritium release

experiments have been carried out in the Japan Materials Testing Reactor (JMTR). Additionally, the effects of various parameters on the tritium release behavior from Li_2TiO_3 pebble beds (i.e. irradiation temperature, sweep gas flow rate) were studied.

2. Experimental

2.1. Irradiation facility

The schematic diagram of the in situ tritium release experiments of a blanket in-pile mockup with a Li_2TiO_3 pebble bed in the JMTR was described in a previous paper [8]. The sweep gas system consists of three main elements: a sweep gas supply system, a tritium measuring system and a tritium recovery system [9]. The sweep gas flow rate in the in-pile mockup can be controlled from 10 to 1000 $\text{N cm}^3/\text{min}$. The hydrogen content in the helium sweep gas can be also controlled from 10 to 10000 ppm H_2 . In the tritium measuring system, the total tritium concentration (HT + HTO) and the gaseous tritium concentration (HT) released from the in-pile mockup can be measured continuously.

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Table 1
 Characteristics of Li_2TiO_3 pebbles fabricated by wet process (7.4% ^6Li -enrichment)

	Large pebble	Small pebble
Fabrication method	Wet process with dehydration reaction	Wet process with substitution reaction
Density	2.86 g/cm ³	2.82 g/cm ³
Sphericity	1.07 (av.)	1.11 (av.)
Diameter	1.7–2.36 mm (≈ 1.9 mm av.)	0.25–0.3 mm (≈ 0.27 mm av.)
Grain size	<5 μm	<5 μm
Collapse load	73.4 N	4.6 N
Impurity	Ca: <2 ppm, Na: 82 ppm, Al: 9 ppm, Si: 14 ppm	Ca: 18 ppm, Na: 39 ppm, Al: 23 ppm, Si: 73 ppm

2.2. Irradiation test with pulse-operation simulating mockup

Two kinds of Li_2TiO_3 pebbles were prepared by the wet process. The large pebbles (diameter ≈ 2 mm) were fabricated by the wet process with the dehydration reaction [10] and the small pebbles (diameter ≈ 0.3 mm) were fabricated by the wet process with the substitution reaction [11]. The characteristics of Li_2TiO_3 pebbles fabricated by wet process is shown in Table 1.

The schematic diagram of the pulse-operation simulating mockup is shown in Fig. 1. The outer diameter of the in-pile mockup was 65 mm which is the available

maximum size in the JMTR. This mockup consists of a hafnium (Hf) neutron absorber to harden the neutron spectrum. The absorber with a window rotated by the stepping motor which was installed in this mockup for pulse-operation simulation of the fusion reactor. Multi-paired thermocouples and self-powered neutron detectors (SPND) for measuring the temperature and the thermal neutron flux and electrical heaters for controlling the irradiation temperature of the Li_2TiO_3 pebble bed were also installed in this mockup. The multi-paired thermocouples (T/Cs) and SPNDs were installed in the in-pile mockup. The dimension of the irradiated binary Li_2TiO_3 pebble bed was 20 mm inner diameter \times 260 mm

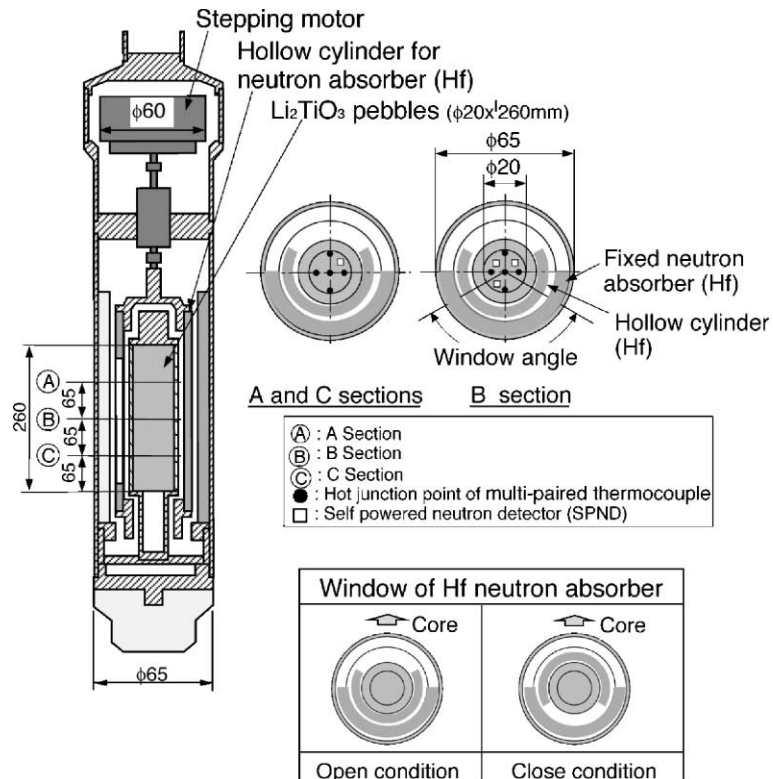


Fig. 1. Schematic diagram of pulse-operation simulating mockup.

Table 2

Main specifications of Li_2TiO_3 pebble bed in the pulse-operation simulating mockup

Packing fraction	81.3%
Loaded weight of large pebbles	129 g
Loaded weight of small pebbles	43 g
Length of packing region	260 mm

length. The main specifications of the Li_2TiO_3 pebble bed are shown in Table 2. The in-pile mockup was irradiated in the irradiation position M-2 of JMTR. Also nuclear calculations were performed in 3-D geometry with the Monte Carlo code MCNP4B using the continuous energy cross-section library FSXLIBJ3R2 (based on JENDL3.2). The calculated result, the tritium generation rate in the Li_2TiO_3 pebble bed, was about 2.3×10^{10} Bq/d when the window of the Hf neutron absorber was opened and about 3.3×10^9 Bq/d when the window of the Hf neutron absorber was closed.

3. Results and discussion

3.1. Tritium release at reactor power-up

The first experiment was conducted in order to evaluate total tritium (HT + HTO) release amount and moisture concentration in the sweep gas at reactor start-up (see Fig. 2). When the outside edge temperature of the Li_2TiO_3 pebble bed became 180 °C, the tritium release started. Then, the release rate of total tritium increased with increasing the outside edge temperature of the Li_2TiO_3 pebble bed. On the other hand, the moisture

concentration in the sweep gas increased up to 250 ppm at the outlet of the Li_2TiO_3 pebble bed. When the outside edge temperature of the Li_2TiO_3 pebble bed was about 300 °C, the moisture concentration was about 20 ppm, the ratio of HT/(HT + HTO) was about 30%. On the other hand, when the outside edge temperature and moisture concentration were about 380 °C and less than 2 ppm, respectively the HT/(HT + HTO) ratio increased to about 70%.

The second experiment was conducted in order to evaluate the effect of irradiation temperature on the tritium release from the Li_2TiO_3 pebble bed. The relationship between the temperature at the outside of the Li_2TiO_3 pebble bed and the (R/G) ratio of the tritium release and generation amounts is shown in Fig. 3: R is a constant value after 2 h. At this time, the sweep gas flow rate and hydrogen content in the sweep gas were 200 cm^3/min and 1000 ppm H_2 , respectively. The moisture concentration was constant and less than 0.1 ppm. When the temperature at the outside edge of the Li_2TiO_3 pebble bed became higher than 100 °C, the tritium release from the bed started. The tritium release amount increased with increasing the temperature at the outside edge of the Li_2TiO_3 pebble bed. From these tests, the R/G ratio was about 1 when the temperature at the outside edge of the Li_2TiO_3 pebble bed became more than 300 °C.

The third experiment was conducted in order to evaluate the effect of the sweep gas flow rate on the tritium release from the Li_2TiO_3 pebble bed. At this time, the hydrogen content in the sweep gas and the temperature at the outside edge of the Li_2TiO_3 pebble bed were 1000 ppm H_2 and about 300 °C, respectively. The moisture concentration was constant and less than

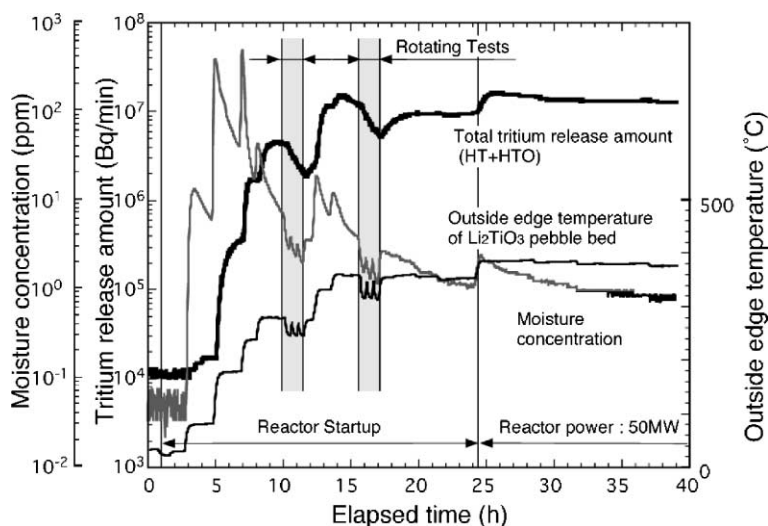


Fig. 2. Result of tritium release at the reactor startup.

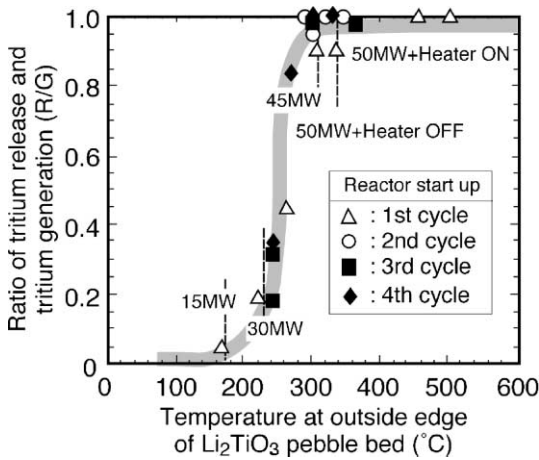


Fig. 3. Relationship between temperature at the outside of the Li_2TiO_3 pebble bed and the (R/G) ratio of the tritium release and tritium generation amounts.

0.1 ppm. In this experiment, when the sweep gas flow rate changed, the tritium release amount changed in a moment. Then, it returned to the same value after about 5 h. The overall rate constant of the tritium desorption was calculated based on the tritium release amount when the sweep gas flow rate changed. This result is shown in Fig. 4: when the amount of the flow rate change increased, it turns out that the overall rate constant of tritium desorption increased. It is considered that the hydrogen content of the surface of the Li_2TiO_3 pebble changed temporarily when the sweep gas flow rate was changed.

The fourth experiment was conducted in order to evaluate the effect of the hydrogen content in the sweep

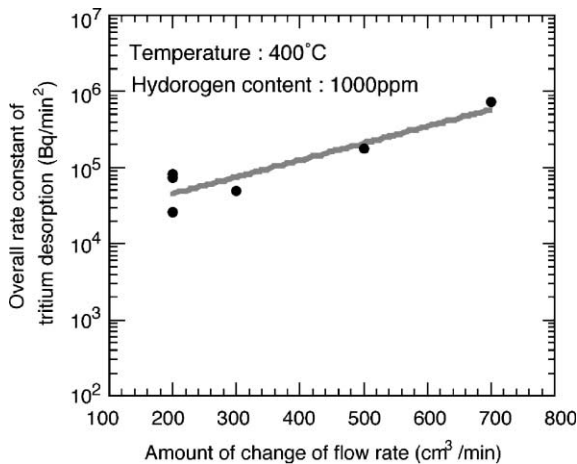


Fig. 4. Relationship between the amount of flow rate change and overall rate constant of tritium desorption.

gas on the tritium release from the Li_2TiO_3 pebble bed. At this time, the sweep gas flow rate and the temperature at the outside edge of the Li_2TiO_3 pebble bed were 200 cm³/min and 300 °C respectively. The moisture concentration was constant and less than 0.1 ppm. The hydrogen content varied from 100 to 10000 ppm H₂. When the hydrogen content increased from 1000 to 10000 ppm H₂, also the tritium release increased. However, the tritium release with 1000 ppm H₂ became the same as that of the tritium release of 10000 ppm H₂ after about 5 h from controlling the hydrogen content. The relationship between the hydrogen content in the sweep gas and the (R/G) ratio of tritium release (R) and tritium generation (G) are shown in Fig. 5: R is constant after more than 5 h. When the hydrogen content was more than 800 ppm H₂, the total tritium release was constant. However, the total tritium release decreased with decreasing hydrogen content when it was less than 800 ppm H₂. In this test, it seems that the effects, such as isotope exchange, surface adsorption, surface desorption and so on, influence the tritium release from the Li_2TiO_3 pebble bed when the hydrogen content was less than 800 ppm H₂.

The tritium release, tritium inventory and so on under ITER pulsed operation with a developed calculation code was reported by Federici et al. [12]. However, tritium release experiments under ITER pulsed operation were not performed. Therefore, the fifth experiment was conducted in order to evaluate the effect of continuous pulsed operation on the tritium release from the Li_2TiO_3 pebble bed. This experiment was carried out under

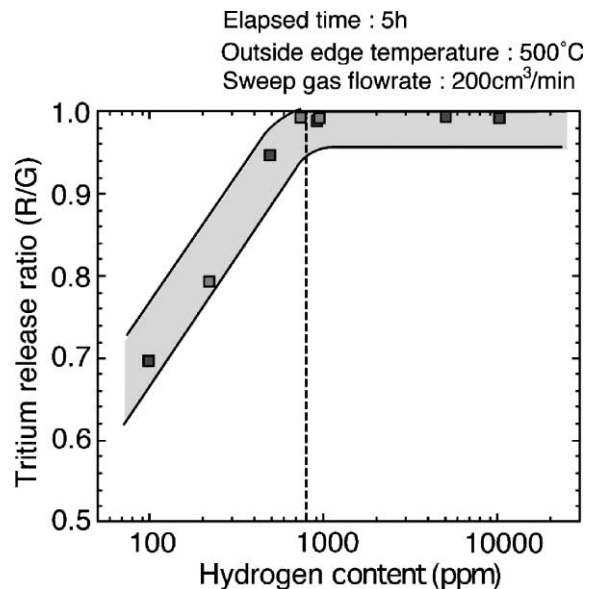


Fig. 5. Relationship between hydrogen content in sweep gas and tritium release (R/G) ratio.

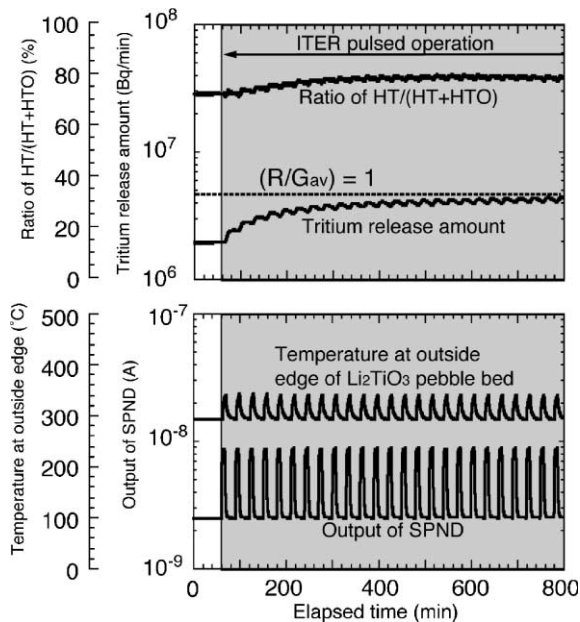


Fig. 6. Result of the rotational test under the condition of ITER pulsed operation.

conditions of the ITER pulsed operation [13]. The periods of open condition and closed condition were 400 and 1310 s, respectively. The number of pulse operation cycles was 200 cycles. The result of this experiment is shown in Fig. 6. At this time, the sweep gas flow rate and hydrogen content in the sweep gas were 400 cm³/min and 1000 ppm H₂, respectively. The moisture concentration was constant and less than 0.1 ppm. Additionally, the (R/G) ratio was about 1 when the window of Hf was closed. In this experiment, the temperature at the outside edge of the Li₂TiO₃ pebble bed changed from about 300–350 °C immediately. On the other hand, the tritium release increased cycle by cycle. After about 20 cycles, the average of the tritium release amount was almost constant and the (R/G_{av}) ratio of the tritium release amount and average tritium generation was saturated, where G_{av} is the average of the tritium generation amount under pulse operation conditions (open time: 400 s, closed time: 1310 s), R is a constant value after more than 20 h. In Ref. [12], the tritium release curve shows a characteristic peak at the beginning of burn time due to the combined effect of the generation rate beginning and the temperature rising. From these results, it is considered that the number of pulse operation cycles when the (R/G_{av}) ratio was saturated, depends on the build-up of the tritium inventory. In future plans, tritium release experiments which control the pulse operation conditions will be performed and the tritium release from the Li₂TiO₃ pebble bed will be evaluated.

4. Conclusion

The effects of irradiation temperature, sweep gas flow rate and hydrogen content on the tritium release behavior of the Li₂TiO₃ pebble bed are summarized as it follows by this in situ tritium release experiments in the JMTR:

- (1) The generated tritium was almost released when the temperature at the outside edge of the Li₂TiO₃ pebble bed became higher than 300 °C. On the other hand, the obtained result suggests that moisture also influences the release rate of total tritium and gaseous tritium at the reactor start-up.
- (2) When the flow rate increases, the overall rate constant of tritium desorption increases. It is considered that the hydrogen content of the surface of the pebble changed temporarily when flow rate was varied.
- (3) The hydrogen content in the sweep gas has an effect on the tritium release from the Li₂TiO₃ pebble bed. The tritium release increased by increase of hydrogen in the sweep gas and decreased by decrease of hydrogen in the sweep gas.
- (4) The tritium release increased cycle by cycle. After about 20 cycles, the average tritium release was almost constant and the (R/G_{av}) ratio has saturated. It is considered that the number of pulse operation cycles, when (R/G_{av}) has saturated, depends on the build-up of the tritium inventory.

From these results of in situ experiments, bright prospects were obtained concerning the design of a breeding blanket with a Li₂TiO₃ pebble bed.

References

- [1] R.A. Verrall, J.M. Miller, L.K. Jones, AECL-11768 (CFFTP G-9637), 1997.
- [2] O.D. Slagle, T. Kurasawa, R.A. Verrall, G.W. Hollenberg, J. Nucl. Mater. 191–194 (1992) 214.
- [3] O.D. Slagle, T. Takahashi, F.D. Hobbs, K. Noda, D.L. Baldwin, G.W. Hollenberg, R.A. Verrall, J. Nucl. Mater. 212–215 (1994) 988.
- [4] R.A. Verrall, O.D. Slagle, G.W. Hollenberg, T. Kurasawa, J.D. Sullivan, J. Nucl. Mater. 212–215 (1994) 902.
- [5] H. Kwast, M. Stijkel, R. Muis, R. Conrad, ECN-C-95-123, 1995.
- [6] J.G. van der Laan, M.P. Stijkel, R. Conrad, JAERI-Conf 98-006, 1998, p. 88.
- [7] J.G. van der Laan, R. Conrad, K. Bakker, N. Roux, M.P. Stijkel, Proceedings of the 20 Symposium on Fusion Technology, France, 1998, p. 1239.
- [8] K. Tsuchiya, M. Nakamichi, Y. Nagao, J. Fujita, H. Sagawa, S. Tanaka, H. Kawamura, Fusion Eng. Des. 51&52 (2000) 887.
- [9] H. Kawamura, H. Sagawa, I. Ishitsuka, K. Tsuchiya, N. Sakamoto, T. Niiho, Proceedings of an ENS Class 1 Topical Meeting, 1996, p. 232.

- [10] K. Tsuchiya, H. Kawamura, K. Fuchinoue, H. Sawada, K. Watarumi, *J. Nucl. Mater.* 258–263 (1998) 1985.
- [11] K. Tsuchiya, H. Kawamura, *J. Nucl. Mater.* 283–287 (2000) 1380.
- [12] G. Federici, C.H. Wu, A.R. Raffray, M.C. Billone, *J. Nucl. Mater.* 187 (1992) 1.
- [13] ITER JCT, ITER-FEAT Outline Design report (ODR), G A0 R12 99-11-22 W 0.1, 1999.