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Breeding Blanket Development

- Tritium Release from Breeder -

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Engineering data on neutron irradiation performance of tritium breeders are needed to design the breeding blanket of fusion reactor. In this study, tritium release experiments of the breeders were carried out to examine the effects of various parameters (such as sweep gas flow rate, hydrogen content in sweep gas, irradiation temperature and thermal neutron flux) on tritium generation and release behavior. Lithium titanate (Li_2TiO_3) is considered as a candidate tritium breeder in the blanket design of International Thermonuclear Experimental Reactor (ITER). As for the shape of the breeder material, a small spherical form is preferred to reduce the thermal stress induced in the breeder. Li_2TiO_3 pebbles of about 170g in total weight and with 0.3 and 2mm in diameter were manufactured by a wet process, and an assembly packed with the binary Li_2TiO_3 pebbles was irradiated in Japan Materials Testing Reactor (JMTR). The tritium was generated in the Li_2TiO_3 pebble bed and released from the pebble bed, and was swept downstream using the sweep gas for on-line analysis of tritium content. Concentration of total tritium and gaseous tritium (HT or T_2 gas) released from the Li_2TiO_3 pebble bed were measured by ionization chambers, and the ratio of (gaseous tritium)/(total tritium) was evaluated. The sweep gas flow rate was changed from 100 to 900cm^3/min, and hydrogen content in the sweep gas was changed from 100 to 10000ppm. Furthermore, thermal neutron flux was changed using a window made of hafnium (Hf) neutron absorber. The irradiation temperature at an outer region of the Li_2TiO_3 pebble bed was held between 200 and 400°C.

The main results of this experiment are summarized as follows.

1) When the temperature at the outside edge of the Li_2TiO_3 pebble bed exceeded 100°C, the tritium release from the Li_2TiO_3 pebble bed started. The ratio of the tritium release rate and the tritium generation rate (normalized tritium release rate : R/G) reached unity when the temperature at the outside edge of the Li_2TiO_3 pebble bed was kept above 300°C.
2) The sweep gas flow rate did not affect the tritium release from the Li_2TiO_3 pebble bed in the steady state condition, when the sweep gas flow rate was changed from 100 to 900cm^3/min (the superficial gas velocity in the Li_2TiO_3 pebble bed : 0.53 - 4.8cm/s).
3) Results of in-situ tests with varied hydrogen contents in the sweep gas showed that the hydrogen partial pressure in the sweep gas had an effect on tritium release from the Li_2TiO_3 pebble bed.

Keywords: ITER, Fusion Reactor, Blanket, Tritium Release, Lithium Titanate, JMTR

This report is based on the final report of the ITER Engineering Design Activities (EDA).

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増殖ブランケット開発
－増殖材料からのトリチウム放出－

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トリチウム増殖材料の中性子照射に関する工学的データの取得は、核融合炉増殖ブランケットの設計に必要不可欠である。スイープガス流量、スイープガス中の水素添加量、照射温度、熱中性子束等によるトリチウム生成・放出に与える影響を評価するためにトリチウム増殖材料のトリチウム放出試験を行った。チタン酸リチウム（Li_2TiO_3）は、熱核融合実験炉（ITER）の増殖ブランケットの候補材料として考えられている。材料中に生じる熱応力の低減の観点から、増殖材料の形状は微小球が提案されている。湿式法により直径0.3mmと2mmの微小球を製造し、約170g充填したLi_2TiO_3微小球充填体を材料試験炉(JMTR)で照射した。トリチウムはLi_2TiO_3微小球充填体中で生成・放出され、スイープガス中のトリチウム量を連続的に測定した。Li_2TiO_3微小球充填体から放出される全トリチウム濃度とガス成分トリチウム濃度を電離箱で測定し、ガス成分と全トリチウムの比を評価した。スイープガス流量は100～900cm^3/minの範囲で、水素添加量は100～10000ppmの範囲で変更した。さらに、ハフニウム（Hf）製中性子吸収体で製作した窓を用いて熱中性子束を変化させた。Li_2TiO_3微小球充填体の外壁温度は200～400℃に保持した。

本試験の主な結果は以下の通りである。

1）Li_2TiO_3微小球充填体の外壁温度が100℃以上になったとき、トリチウム放出が観測された。また、充填体の外壁温度を300℃以上に保持したとき、トリチウム放出率（R/G）は1に到達した。

2）スイープガス流量を100～900cm^3/min（Li_2TiO_3微小球充填体の空塔速度：0.53～4.8cm/s）の範囲で変化させても、定常時におけるLi_2TiO_3微小球充填体からのトリチウム放出に影響はなかった。

3）スイープガス中の水素添加量はトリチウム放出に影響があることが分かった。

本報告書は、ITER工学設計報告書に補筆を行ったものである。

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1.1 Background and Objective

Engineering data on neutron irradiation performance of tritium breeder are needed to design the breeding blanket of fusion reactor. Knowledge about the performance of solid lithium-based ceramic is limited at present. Tritium will be bred in the blanket surrounding the plasma in the vacuum vessel, and will be collected and injected into the plasma chamber as fuel.

In the development of tritium breeding blankets for fusion reactors, lithium-based ceramics such as Li_2O, Li_2TiO_3, Li_2ZrO_3, $LiAlO_2$ and Li_4SiO_4 have been recognized as promising tritium breeding materials [1-2]. Particularly, Li_2TiO_3 has attracted the attention of many researchers from viewpoints of easy tritium release at low temperatures, chemical stability at the high temperatures, etc. [3-5]. Application of small lithium based ceramic pebble was proposed in the breeding blanket design in order to reduce thermal stress [6-9].

Li_2TiO_3 pebbles of about 170g in total weight and with 0.3 and 2mm in diameter were manufactured by a wet process, and an assembly packed with the binary Li_2TiO_3 pebbles was irradiated in Japan Materials Testing Reactor (JMTR) for 3 cycles (about 75 day). The tritium generated in the Li_2TiO_3 pebble bed, was swept downstream by the sweep gas for on-line analysis of tritium content.

In this study, tritium release experiments of the breeders were carried out to examine the effects of various parameters (such as sweep gas flow rate, hydrogen content in sweep gas, irradiation temperature and thermal neutron flux) on tritium generation and release behavior. Some papers and reports were published so far on research and development for these experiments and on analyzed results of these in-situ experiments [10-15].

1.2 Task Description

In-situ data on tritium release from tritium breeding material at low temperature are indispensable for the design of the ITER breeding blanket. An irradiation test for the task (ITER Task No. : T431) was planned to be carried out by using JMTR under transient neutron irradiation conditions to simulate the ITER pulse operation. The irradiation facility for in-situ irradiation test is shown in Fig.1.

The tritium release could be varied by changing the thermal neutron flux by rotating a window made of hafnium. The test condition in this task is shown in the following table.

Test Conditions in This Task (T-431)

Irradiation Capsule	
- Shape	see Fig.2
- Packing Weight of Tritium Breeding Material	about 170 g
- Packing Fraction of Tritium Breeding Material	about 80%
- Structural Material	SS316
- Neutron Absorber	Hafnium (Hf)
Tritium Breeding Material	
- Material	Li_2TiO_3
- Shape	Pebble
- Diameter	about 0.3 and 2mm
- Density	about 83 %T.D.
- ^6Li Enrichment	natural
- Grain Size	about 5μm
Instrumentation (Number)	
- Multi-Paired Thermocouple	5 (3 hot junctions /15measurement points)
- Self-Powered Neutron Detector (SPND)	5
Irradiation Conditions	
- Reactor	JMTR
- Irradiation Hole	M-2
- Irradiation Period	3 cycles in JMTR(1 cycle ≅ 25 days)
- Neutron Flux	
Thermal Neutron (E<0.683eV)	3×10^{15} - 2×10^{16} n/m^2/s (TBD*)
Fast Neutron (E>1.0 MeV)	5×10^{15} n/m^2/s (TBD*)
- Tritium Generation Rate	1.85×10^{11} Bq/d (TBD*)
- Irradiation Temperature (Tritium Breeder Region)	200 - 400 °C
- Volumetric Heating Rate	2 W/cm^3 (TBD*)
- Changing Width of Thermal Neutron Flux	
Closed Condition of Hf Window	3×10^{15} n/m^2/s (TBD*)
Open Condition of Hf Window	2×10^{16} n/m^2/s (TBD*)
Sweep Gas Conditions	
- Sweep Gas Composition	pure He, He+H_2
- Sweep Gas Flow Rate	100 - 900cm^3/min
- Hydrogen Content in Sweep Gas	100 - 10000ppm
Tritium Release Test	
- Change of Sweep Gas Flow Rate	
- Change of Hydrogen Content in Sweep Gas	
- Change of Irradiation Temperature and Neutron Flux	

(* TBD : to be detected)

2. Preparation of Li_2TiO_3 Pebbles

Two kinds of Li_2TiO_3 pebbles were fabricated by the wet process with the dehydration reaction [16] and the substitution reaction [17]. Furthermore, characteristics of these Li_2TiO_3 pebbles were examined.

2.1 Fabrication of Li_2TiO_3 Powder

Li_2CO_3 and TiO_2 powders, which are starting materials, were prepared with purity of 99.99% and 99.9%, respectively. The Li_2TiO_3 powder was prepared by a solid state reaction. The reaction for production of Li_2TiO_3 powder is expressed by Eq.(1).

$$Li_2CO_3 + TiO_2 \rightarrow Li_2TiO_3 + CO_2 \qquad (1)$$

Mixed powder (Li_2CO_3 and TiO_2) was pulverized and reacted in air at 700~800°C for 8 h. After the reaction, the produced Li_2TiO_3 powder was pulverized, and particle size of the Li_2TiO_3 powder was measured by a laser diffraction method. Impurities in the Li_2TiO_3 powder were measured by an atomic emission spectrometer with inductively coupled plasma (ICP-AES) and an atomic absorption spectorometer (AAS).

The particle size of the Li_2TiO_3 powder was in a range of 0.2-2.3 μm, and was 0.63 μm on average. Impurities of the Li_2TiO_3 powder are shown in Table 1. Silicon (Si) and sodium (Na) were the highest impurities detected in the Li_2TiO_3 powder. Crystal structure of this powder was measured by X-ray diffractometry (XRD). The X-ray diffraction pattern of the Li_2TiO_3 powder is shown in Fig.3, and Li_2TiO_3 was the main component detected.

2.2 Fabrication of Li_2TiO_3 Pebbles

The fabrication flow of Li_2TiO_3 pebbles for the wet process with the dehydration reaction and the substitution reaction is shown in Fig.4. These procedures include fabrication of gel-spheres, calcination and sintering processes.

The large pebbles (diameter : about 2mm) were fabricated by the wet process with the dehydration reaction and the main processes [18] is described as follows;

1) Fabrication of gel-spheres: Mixed slurry of Li_2TiO_3 powder and polyvinylalcohol (PVA) solution was dropped into cooled acetone (-30°C) through a nozzle, and gel-spheres were fabricated.

2) Calcination of gel-spheres: The PVA in the gel-spheres was removed in air at 600°C for 6 h, and Li_2TiO_3 pebbles with a low density are fabricated in this process.
3) Sintering: Li_2TiO_3 pebbles with a low density were sintered in air at 1200°C for 15 minutes, and Li_2TiO_3 pebbles with a high density were fabricated.

The small pebbles (diameter : about 0.3mm) were fabricated by the wet process with the substitution reaction, and the main processes [18] is described as follows;

1) Fabrication of gel-spheres: A liquid mixture of Li_2TiO_3 powder and sodium alginate solution as the binder was dropped into an aqueous solution of zinc chloride through a nozzle, and gel-spheres were generated.
2) Calcination of gel-spheres: Zinc in the gel-spheres was removed in the Ar+H_2 gas atmosphere at 800°C for 8 h, and Li_2TiO_3 pebbles with a low density were obtained.
3) Sintering: The Li_2TiO_3 pebbles were sintered in air at 1200°C for 15 minutes, and Li_2TiO_3 pebbles with a high density were fabricated.

Each gel-sphere was fabricated by the automatic dropping system. A schematic drawing of this system for fabrication of gel-spheres is shown in Fig.5. This system consists of a slurry tank, a vibration generator, a nozzle for dropping and a solution tank for aging. The diameter of droplets is given as follows:

$$D = \left(\frac{6 \cdot Q}{\pi \cdot f}\right)^{\frac{1}{3}}, \qquad (2)$$

where D (cm) is the diameter, Q (cm^3/s) is the flow rate of the liquid mixture, and f (Hz) is the frequency of nozzle. The frequency of the vibration generator is given by the oscillator. On the other hand, the diameter of the droplets is influenced by the viscosity of the liquid mixture, the nozzle diameter, and wettability between the nozzle and the liquid mixture. Thus, the optimum flow rate and frequency were decided for the fabrication of gel-spheres with a target diameter. Additionally, the sphericity was also influenced by the viscosity of the liquid mixture and the frequency of the nozzle.

2.3 Characterization of Li_2TiO_3 Pebbles

Characteristics of the two kinds of Li_2TiO_3 pebbles fabricated are summarized in Table 2. The main features are discussed below.

Photographs of Li_2TiO_3 pebbles fabricated by the wet process described above are shown in Fig.6. Diameter distribution of the two kinds of Li_2TiO_3 pebbles is shown in Fig.7. Average diameters of these two kinds of Li_2TiO_3 pebbles were 1.93mm and 0.29mm, respectively. Sphericity (defined as the ratio of the largest diameter to the smallest diameter of each pebble) of the two kinds of Li_2TiO_3 pebbles was measured by a photographic analysis method. The sphericity was as large as 1.05 - 1.1.

SEM photographs of cross sections of the two kinds of Li_2TiO_3 pebbles are shown in Fig.8. The grain size was measured by using these photographs of the cross section. The average grain size of two kinds of Li_2TiO_3 pebbles was smaller than 5μm. The density of the two kinds of Li_2TiO_3 pebbles was measured by mercury porosimetry, and the average densities were 2.86 and 2.82g/cm^3 for the large and small pebbles, respectively.

Results of impurity measurements of the two kinds of Li_2TiO_3 pebbles are shown in Table 3. Sodium (Na) and carbon (C) were the highest impurities detected in the large pebbles. On the other hand, aluminum (Al), silicon (Si), sodium (Na) and iron (Fe) were the highest impurities detected in the small pebbles. It seems that these impurities were introduced from each binder material (namely, PVA and ammonium alginate).

To evaluate the strength of Li_2TiO_3 pebbles fabricated by these methods, the crushing strength was measured by a compression strength test. The average crushing load of the large and small pebbles was about 73 and 4.6 N, respectively.

An X-ray diffraction pattern of the Li_2TiO_3 pebbles is shown in Fig.9. The XRD analysis of the two kinds of Li_2TiO_3 pebbles was undertaken after packing the pebbles in a polyethylene sheet. In the same manner as in Fig.3, Li_2TiO_3 was the main component detected.

3. Irradiation Capsule and Facility

3.1 Fabrication of Irradiation Capsule

A vertical cross-section of the irradiation capsule (capsule name in JMTR : 99M-54J) is shown in Fig.10. The outer diameter of capsule is 65mm which is the maximum available size in JMTR. The capsule was equipped with thermocouples and SPNDs, mentioned below, to measure the temperature and thermal neutron flux, respectively, and with electrical heaters to control the irradiation temperature of the tritium breeding material. Furthermore, thermal neutron flux could be changed by rotating a window made of hafnium around the inner capsule [11]. The photograph of the inner capsule to be installed in the capsule is shown in Fig.11. The inner capsule is made of

SS316L, and the dimension of the Li_2TiO_3 pebble container is $23mm^{OD}\times 20mm^{ID}\times 360mm^{L}$. A binary Li_2TiO_3 pebble bed was fabricated using the inner capsule.

(1) Packing fraction and effective thermal conductivity

Results of packing fraction measurements of Li_2TiO_3 pebbles are shown in Fig.12. Main features of the pebble bed are as follows.

Li_2TiO_3 pebble region

- Packing fraction: 81.4%
- Loaded weight: 171.81 g
- Length of packing region: 261.2 mm

A calculation result of effective thermal conductivity of the binary Li_2TiO_3 pebble bed is shown in Fig.13. The calculation was performed in the following supposition.

-Packing fraction : 80% (Large pebbles = 63%, Small pebbles = 17%)
- Li_2TiO_3 pebble diameter : Large pebbles = ϕ2mm, Small pebbles = ϕ0.3mm
- Li_2TiO_3 pebble density : 83%T.D.
- Li_2TiO_3 thermal conductivity : Equation by Saito et al. [19]
-Effective thermal conductivity in binary packing : equation by SZB [20]
(SZB : Schlunder, Zehner and Bauer)

(2) Instruments in inner capsule

Two kinds of instruments, i.e. multi-paired thermocouples (T/C) and self-powered neutron detector (SPND) were used in this inner capsule. Arrangement of the multi-paired thermocouples and SPNDs at sections A, B and C is shown in Fig.14. The vertical positions of sections A, B and C in Fig.14 are 75, 130 and 205mm, respectively, from the vertical top end of Li_2TiO_3 pebble packing region. At section A, there are 5 T/Cs and one SPND; and at section B, 5 T/Cs and 3 SPNDs; at section C, 5 T/Cs and one SPND. Conceptual structures of the multi-paired thermocouple and the self-powered neutron detector are described in Figs.15 and 16, respectively.

As for the thermocouples, the multi-paired thermocouples with three hot junctions were used. This was because a lot of thermocouples were necessary from a viewpoint of exact measurement of the irradiation temperature distribution, while the numbers of instruments through the upper plug of the inner capsule were limited due to difficulty in micro-brazing. The outer diameter of the multi-paired thermocouple was 1.8mm, and three small thermocouples were included in one multi-paired thermocouple. The outer diameter of the small thermocouple was 0.5mm. The vertical position of hot junctions of each small thermocouple were 75, 130 and 205mm (namely, the positions of sections A, B and C in Fig.14, respectively) from the vertical top end of the Li_2TiO_3 pebble packing region. For example, the irradiation temperature distribution in the Li_2TiO_3 pebble packing region at section B were measured by the use of thermocouples #6-#10 (see Fig.14).

The emitter of SPND was Rh-60%Pt, and the outer diameters of the corrector and the cable were 2mm and 1.5mm, respectively. Additionally, the irradiation temperature in the Li_2TiO_3 pebble packing region was controlled by a micro-heater instrumented at the outside of the inner capsule. The capacity of the heater was 2kW at maximum.

(3) Loading position and nuclear/thermal calculations

This capsule was irradiated in the irradiation hole M-2 in JMTR. The core configuration of JMTR is shown in Fig.17. This irradiation hole is located at the outside of a γ-ray shielding plate made of Zr. This hole was the most convenient for conducting an irradiation test of tritium breeder because of a minimum effect of α-heating by (n, α) reaction and γ-heating of stainless steel, compared with heating by the mico-heater.

A conceptual structure of the in-pile mockup and a nuclear calculation model of the JMTR core are shown in Fig.18. Nuclear calculations with a Monte Carlo code MCNP [21] revealed α-heat and γ-heat as follows.

[Open condition of Hf window]
α-heat : 0.754 W/g
γ-heat : 0.165 W/g
[Closed condition of Hf window]
α-heat : 0.110 W/g
γ-heat : 0.140 W/g

A calculation model of the irradiation temperature distribution in the Li_2TiO_3 pebble bed is shown in Fig.19, and the calculation parameters are given in Table 4. A result of the nuclear and thermal calculations of irradiation capsule is shown in Fig.20. For the full power (50MW) operation of JMTR, the centerline temperature of the Li_2TiO_3 pebble packing region was about

356°C without the micro-heater when the Hf window was opened. Calculation results for the in-pile mockup by MCNP code and GENGTC code [22] are given in Table 5.

3.2 Irradiation Facility for In-situ Irradiation Test

Irradiation facility for the in-situ irradiation test was described in references [10, 23] in detail.

The tritium measurement system in the facility consists of four subsystems: a sweep gas supply subsystem, a tritium measuring subsystem, a tritium recovery subsystem and a clean up subsystem of an operation box with gloves. The block diagram of these subsystems and the schematic flow diagram of the sweep gas system were also given in ref. [23].

Results of a design study showed that the sweep gas system is sufficient to support the in-pile functional test. The sweep gas supply subsystem can change broadly the sweep-gas flow rate and the hydrogen content in the sweep gas. In the tritium measuring subsystem, accurate measurement can be made without being concerned about the increase of the background by installing two ceramic electrolytic cells in series. In the tritium recovery subsystem, the exhaust tritium would be less than 1×10^{10} Bq/y. Even if an accident such as a piping rupture occurs, the exposure would be minimized by the clean up subsystem.

4. Results of Irradiation Tests

4.1 Results of Irradiation Test in The 136th Cycle of JMTR

4.1.1 Outline of the First Irradiation Test

Tritium was generated in the Li_2TiO_3 pebble bed by neutron irradiation, and released from the Li_2TiO_3 pebble bed. The Li_2TiO_3 pebble bed was swept by He gas and the tritium was released from the Li_2TiO_3 pebble bed. Total tritium concentration (HT+HTO) and gaseous tritium concentration (HT) were measured separately, and HT/(HT+HTO) ratio was evaluated.

The first irradiation test of the Li_2TiO_3 pebble bed with JMTR was conducted from November 17 to December 12, 2000 at the 50MW full power. The outline of experimental conditions in the first irradiation test is shown in Fig.21. Specially, the first irradiation test was focused on low-temperature irradiation behavior of the Li_2TiO_3 pebble bed. Therefore, the center temperature of Li_2TiO_3 pebble bed was kept at about 400°C basically, and was increased to about 550°C by heating with the heater installed in the irradiation capsule when the Hf window was opened. On the other hand, the center temperature of the Li_2TiO_3 pebble bed was about 330°C when the Hf window was closed. The sweep gas flow rate was kept constant at 200cm^3/min. The hydrogen content in the sweep gas was also constant at 1000ppm.

It has been considered that the moisture in the Li_2TiO_3 pebbles also affects the tritium release behavior from the Li_2TiO_3 pebble bed. On the other hand, the test blanket module for ITER does not have a heating device for preheating the tritium breeder region before neutron irradiation and a lot of moisture will be released from the Li_2TiO_3 pebble bed. Therefore, the effects of moisture concentration at the capsule outlet on tritium release behavior were investigated with this irradiation test. Especially, the packing of Li_2TiO_3 pebbles installed in the inner capsule was carried out in the grove box, and moisture in the grove box was measured and controlled during the packing of the pebbles.

The result of the first irradiation test is shown in Fig.22. Results concerning the effects of the irradiation temperature and the change of thermal neutron flux on tritium release from the Li_2TiO_3 pebble bed were obtained. These are described as follows in turn.

4.1.2 Temperature Distribution

The vertical temperature distribution in the inner capsule at 50 MW is shown in Fig.23. The vertical temperature distribution was approximately uniform within the Li_2TiO_3 packing region (260 mm in height), irrespective of the open or closed condition of the Hf window.

4.1.3 Tritium Release at Reactor Power-up

Figure 24 shows the release rate of total tritium (HT+HTO), the moisture concentration at the capsule outlet, and the center temperature measured by thermocouple #7 at the reactor power-up. When the reactor power became 10MW, the center temperature measured by thermocouple #7 became about 180°C (the Hf window was opened). The tritium release from the Li_2TiO_3 pebble bed began to be observed at the reactor power of 10MW. Then, the release rate of the total tritium increased with increasing the irradiation temperature of the Li_2TiO_3 pebble bed. The moisture concentration in the sweep gas also increased up to 250ppm at 10MW.

When the reactor power became 30MW, the center temperature measured by thermocouple #7 was 292°C, and the moisture concentration at the capsule outlet was 20ppm. At this time, the ratio of HT/(HT+HTO) was about 30%. Furthermore, when the reactor power became 50MW, the ratio of HT/(HT+HTO) increased to about 70%. It is clear that the ratio of HT/(HT+HTO) increased with decreasing the moisture concentration. Finally, the ratio of HT/(HT+HTO) became about 75% after operation at 50MW for more than 10h.

4.1.4 Rotating Tests of Hf Window

Hf window rotating tests were conducted at reactor powers of 30, 40 and 50MW, in order to examine the effect of the changes of the thermal neutron flux and the irradiation temperature on tritium release from the Li_2TiO_3 pebble bed. The condition of the ITER pulse operation is shown in Fig.25, where the open time of the Hf window is 400 s and the closed time is 1310 s. The sequence was adopted in this experiment.

Results of the rotating tests at 30, 45 and 50MW are shown in Figs.26, 27 and 28, respectively. When the rotating test was carried out, the release rate of tritium decreased all over. However, the release rate of tritium was not changed immediately. On the other hand, the moisture concentration was changed rather rapidly with rotation of the Hf window. The outputs of thermocouples and SPNDs were also changed immediately with rotation of the Hf window.

4.1.5 Change of Moisture Concentration at Capsule Outlet

The change of moisture concentration was measured at the capsule outlet on tritium release from the Li_2TiO_3 pebble bed at the reactor start-up. The sweep gas flow rate was 200cm^3/min, and the maximum moisture concentration in the sweep gas at the capsule outlet was about 600ppm. When the reactor power reached just 50MW, the center temperature in the Li_2TiO_3 packing region measured by thermocouple #7 was about 400°C, and the moisture concentration at the capsule outlet was about 0.5 ppm.

4.2 Results of Irradiation Test in The 137th Cycle of JMTR

4.2.1 Outline of the Second Irradiation Test

The second irradiation test of the Li_2TiO_3 pebble bed was conducted from January 12 to February 6, 2001 at the 50MW full power. Outline of experimental conditions in the second irradiation test is shown in Fig.29. This test was started with the hafnium window closed. This test was focused on tritium release behavior from the Li_2TiO_3 pebble-bed when the hafnium window was rotating continuously and when the sweep gas flow rate was changed. Therefore, the center temperature of Li_2TiO_3 pebble bed was about 400°C basically, and was increased to about 550°C by using the micro-heater installed in the irradiation capsule when the Hf window was opened. On the other hand, the center temperature of the Li_2TiO_3 pebble bed was about 330°C when the Hf window was closed. The hydrogen concentration in sweep gas was constant at 1000ppm.

The result of the second irradiation test is shown in Fig.30. This figure gives the results concerning the effects of the irradiation temperature, the change of the thermal neutron flux and the change of the sweep-gas flow rate on tritium release from the Li_2TiO_3 pebble bed. These are described as follows.

4.2.2 Tritium Release at Reactor Power-up

Figure 31 shows the release rate of total tritium (HT+HTO) and gaseous tritium (HT), the moisture concentration at the capsule outlet and the center temperature measured by thermocouple #7 at the reactor power-up. When the reactor power became 10MW, the center temperature (thermocouple #7) became about 170°C when the Hf window was closed. And the tritium release from the Li_2TiO_3 pebble bed began to be observed at 10MW. Then, the release rate of total tritium increased with increasing the irradiation temperature of the Li_2TiO_3 pebble bed. The moisture concentration in the sweep gas also increased up to 40ppm at 10MW. At this time, the ratio of HT/(HT+HTO) was about 3%.

When the reactor power became 15MW, the center temperature measured by thermocouple #7 was 223°C, and the moisture concentration of capsule outlet was about 3ppm. At this time, the ratio of HT/(HT+HTO) was about 5%. The ratio of HT/(HT+HTO) increased with decreasing the moisture concentration. Additionally, when the reactor power became 45MW (the center temperature: 327°C), the ratio of HT/(HT+HTO) increased to about 80%.

4.2.3 Rotating Tests of Hf Window

The rotating tests were conducted at reactor powers of 30 and 45MW in the same time sequence as in Fig.25.

Results of the rotating test at 30 and 45 MW are shown in Figs.32 and 33, respectively. Almost the same results as in 4.1.4 were obtained.

4.2.4 Continuous Rotating Tests of Hf Window

The continuous rotating tests were conducted at 50MW. The test conditions (Run 1 & Run 2) of the ITER pulse operation are shown in Fig.34. The tests of the ITER pulse operation were continuously made up to 200 pulses.

The result of the continuous rotating test at 50MW for Run 1 is shown in Fig.35, and enlarged figures at the start of Hf window rotation (Run 1-1) and after about 100 pulses (Run 1-2) for Run 1 are shown in Figs.36 and 37, respectively. On the other hand, the result of the continuous rotating test at 50MW for Run 2 is shown in Fig.38, and enlarged figures in the start of Hf window rotation (Run 2-1), after about 100 pulses (Run 2-2) and after stopping the Hf window rotation (Run 2-3) for Run 2 are shown in Figs.39, 40 and 41, respectively.

When the Hf window was opened or closed, the outputs of thermocouples and SPNDs were changed immediately in these tests. The release rate of tritium was not changed immediately, but the release rate was increased wavelike. In Run 1-1, the release rate of tritium increased with increasing the number of the pulses up to about 20. The release rate of tritium decreased with increasing the number of the pulses up to about 20 in Run 2-1. The moisture concentration was almost unchanged for rotation of the Hf window.

4.2.5 Sweep Gas Flow Rate Changing Tests

Results of the tests are shown in Fig.42. The sweep gas flow rate was changed as follows: from 200 to 900cm^3/min (Test 1), from 900 to 600cm^3/min (Test 2), from 600 to 100cm^3/min (Test 3), from 100 to 400cm^3/min (Test 4) and from 400 to 200cm^3/min (Test 5).

As seen in Fig.42, the change of the sweep gas flow rates was followed by positive tritium release peaks for the increase of the sweep gas flow rates (Tests 1 and 4), and by negative release peaks for the decrease in the sweep gas flow rates (Tests 2, 3 and 5). However, after about 5 h from the time of the flow rate change, the tritium release rate returned to that before changing the sweep gas flow rate.

4.3 Results of Irradiation Test in The 138th Cycle of JMTR

4.3.1 Outline of the Third Irradiation Test

The third irradiation test of Li_2TiO_3 pebble bed with JMTR was conducted from February 26 to March 23, 2001 at the 50MW full power. Outline of experimental conditions on the third irradiation test is shown in Fig.43. This test was made for the same objective and in almost the same conditions as described in 4.2.1. The center temperature of Li_2TiO_3 pebble bed was about 350°C when the Hf window was closed. The result of the third irradiation test is shown in Fig.44. These are described as follows.

4.3.2 Tritium Release at Reactor Power-up

Figure 45 shows the release rate of total tritium (HT+HTO) and gaseous tritium (HT), moisture concentration at the capsule outlet and the center temperature measured by thermocouple #7 at the reactor power-up. When the reactor power became 10 MW, the center temperature (thermocouple #7) became about 170°C when the Hf window was closed. The tritium release from the Li_2TiO_3 pebble-bed began to be observed at 10MW. Then, the release rate of total tritium increased with increasing the irradiation temperature of Li_2TiO_3 pebble bed. The moisture concentration in the sweep gas also increased up to 1.5ppm. At this time, the ratio of HT/(HT+HTO) was about 3%.

When the reactor power became 15MW, the center temperature measured by thermocouple #7 was 208°C, and the moisture concentration at the capsule outlet was about 0.15ppm. At this time, the ratio of HT/(HT+HTO) was about 5%. The ratio of HT/(HT+HTO) increased with decreasing the moisture concentration. Additionally, when the reactor power became 50MW (the center temperature : 347°C), the ratio of HT/(HT+HTO) increased to about 70%.

4.3.3 Continuous Rotating Tests of Hf Window

The continuous rotating tests were conducted at 50MW. The test conditions (Runs 1 and 2) of the ITER pulse operation were the same as shown above in Fig.34. The tests were continuously made up to 100 pulses.

The result of the continuous rotating test at the reactor power of 50MW for Run 1 is shown in Fig.46, and enlarged figures at the start of Hf window rotation (Run 1-1), after about 50 pulses (Run 1-2) and after stopping the Hf window rotation (Run 1-3) for Run 1 are shown in Figs.47, 48 and 49, respectively. On the other hand, the result of the continuous rotating test at the reactor power of 50MW for Run 2 is shown in Fig.50, and detailed figures at the start of Hf window rotation (Run 2-1), after about 50 pulses (Run 2-2) and after stop the Hf window rotation (Run 2-3) for Run 2 are shown in Figs.51, 52 and 53, respectively.

Almost the same results as in 4.2.4 were obtained.

4.3.4 Hydrogen Content Changing Tests

The hydrogen content in the sweep gas was changed as follows: from 1000 to 100ppm (Test 1), from 100 to 1000ppm (Test 2), from 1000 to 10000ppm (Test 3), from 10000 to 1000ppm (Test 4), from 1000 to 5000ppm (Test 5), from 5000 to 500 ppm (Test 6) and from 500 to 1000ppm (Test 7).

The results of these tests are shown in Fig.54. When the hydrogen content was decreased from 1000 to 100ppm (Test 1) and 5000 to 500ppm (Test 6), the tritium release rate also decreased. After about 24 h from the time of the hydrogen content change, the tritium release rate was smaller than that at the hydrogen content of 1000ppm, and became constant. Additionally, the ratio of HT/(HT+HTO) was decreased down to 60%. When the hydrogen content was increased from 1000 to 10000ppm (Test 2) and from 1000 to 5000ppm (Test 5), the tritium release rate increased. After about 15 h from the time of the hydrogen content change, the tritium release rate was almost the same as that before the hydrogen content change and became constant.

5. Discussion

5.1 Summary of General Results

The tritium release from Li_2TiO_3 pebble bed was evaluated when the Hf window was opened or closed. Results of tritium release from the Li_2TiO_3 pebble bed when the window turned toward the reactor core (open condition) and the opposite direction of reactor core (close condition) are shown in Fig.55. At this time, the periods of open and close conditions of the Hf neutron absorber window were 6 and 18 h, respectively. The sweep gas flow rate and the hydrogen content in the sweep gas were 200cm^3/min and 1000ppm, respectively. The normalized tritium release rate (R/G) is the ratio of tritium release rate (R) and tritium generation rate (G) corrected with values calculated by MCNP code.

Figure 55 shows that the center temperature of the Li_2TiO_3 pebble bed increased from 400 to 450°C immediately, when the Hf window was opened. The tritium release rate increased cycle by cycle and $(R/G)_{open}$, i.e. R/G at the open condition approached unity generally. On the other hand, the center temperature of the Li_2TiO_3 pebble bed decreased from 450 to 400°C immediately, when the Hf window was closed. The tritium release rate decreased cycle by cycle, and $(R/G)_{close}$ approached unity. The time constants for the changes in the irradiation temperature and the tritium release were about 100 and 18000s [12], respectively.

5.2 Effect of Continuous Pulsed Operation

The effect of continuous pulsed operation on tritium release from the Li_2TiO_3 pebble bed is discussed here. These experiments were carried out in the conditions of the nominal pulsed operation of ITER. The periods of the open and close conditions are 400 and 1310 s, respectively. The number of the pulse operation cycles was 200.

Results of these experiments are shown in Fig.56. At this time, the sweep gas flow rate and the hydrogen content in the sweep gas were 200cm^3/min and 1000ppm, respectively. Especially, $(R/G)_{close}$ was about unity when the Hf window was closed. In this experiment, the center temperature of the Li_2TiO_3 pebble bed changed from 350 to 400°C immediately, and outputs of SPNDs also changed in a moment.

On the other hand, the tritium release rate increased cycle by cycle. After about 20 pulses, the average of the tritium release rate was almost constant and the normalized tritium release rate

$(R/G)_{av}$ approached unity, where G is the average of tritium generation rate under the pulse operation conditions (open condition : 400s, close condition : 1310s). Thus, it is found from these tests that the number of pulse operation cycles to obtain unity for $(R/G)_{av}$, depends on the pulse operation condition and the irradiation temperature.

5.3 Effect of Irradiation Temperature

Figure 57 summarizes relationship between the normalized tritium release rate (R/G) and the temperature at the outside edge of the Li_2TiO_3 pebble bed, i.e. the lowest temperature in the pebble bed. At this time, the sweep-gas flow rate and the hydrogen content in the sweep gas were 200cm^3/min and 1000ppm, respectively. When the temperature at the outside edge of the Li_2TiO_3 pebble bed exceeded 100°C, the tritium release from the Li_2TiO_3 pebble bed began to be observed. The tritium release rate increased with increasing the temperature at the outside edge of the Li_2TiO_3 pebble bed. These results showed that the normalized tritium release rate (R/G) was about unity when the temperature at the outside edge of the Li_2TiO_3 pebble bed was kept above 300°C.

5.4 Effect of Sweep Gas Flow Rate

In this study, the effect of sweep-gas flow rate on tritium release from the Li_2TiO_3 pebble bed was examined with a fixed value of 1000ppm for the hydrogen content in the sweep gas. When the sweep gas flow rate was increased from 200 to 900cm^3/min, the tritium release rate increased in a moment. However, the tritium release rate returned to that before changing the sweep gas flow rate, after about 5 h from the time of the sweep gas flow rate change.

When the sweep gas flow rate was decreased from 900 to 600cm^3/min, the tritium release rate decreased in a moment. After about 5 h from the time of the sweep gas flow rate change, the tritium release rate returned to that before changing the sweep gas flow rate.

Results of these tests are summarized in Fig.58. This figure shows that the tritium release rate is constant and the normalized tritium release rate (R/G) is about unity for the sweep gas flow rate from 100 to 900cm^3/min (the superficial gas velocity in the Li_2TiO_3 pebble bed : 0.53 - 4.8 cm/s).

5.5 Effect of Hydrogen Content in the Sweep Gas

The effect of hydrogen content in the sweep gas on tritium release from Li_2TiO_3 pebble bed was examined in this study under the conditions that the center temperature of Li_2TiO_3 pebble bed was fixed at 400°C and the sweep gas flow rate was 200cm^3/min.

The hydrogen content was changed from 100 to 10000ppm. For example, when the hydrogen content was changed from 1000 to 10000ppm, the tritium release rate increased. However, the tritium release rate returned to that before changing the hydrogen content, after about 5 h from the time of the hydrogen content change.

Relationship between the hydrogen partial pressure in the sweep gas and the normalized tritium release rate (R/G) is summarized in Fig.59. When the hydrogen partial pressure was higher than 1 Pa, the total tritium release rate was constant. However, the total tritium release rate decreased with decreasing the hydrogen partial pressure when the hydrogen partial pressure was lower than 1 Pa. These results showed that the hydrogen partial pressure in the sweep gas had an effect on tritium release from the Li_2TiO_3 pebble-bed.

6. Conclusions

The in-situ irradiation test with the Li_2TiO_3 pebble bed was started using JMTR, and tritium release characteristics were examined at reactor start-up by using the in-situ measuring system. The particular implications of the results are as follows:

1) The release rate of total tritium increased with increasing the temperature at the center of the Li_2TiO_3 pebble bed. When the reactor power reached 50MW, the temperature at the outer region of the Li_2TiO_3 pebble-bed was at about 350°C, and the release rate of total tritium and gaseous tritium was about 1.6×10^7 and about 1.1×10^7 Bq/min, respectively. When the reactor power reached 30 MW in the first irradiation test, the ratio of HT/(HT+HTO) was about 30%. The ratio of HT/(HT+HTO) increased with decreasing the moisture concentration. The relative amount of HT increased to about 75% of the total tritium. These results show that the moisture concentration as well as the temperature influenced the release rate of total tritium and gaseous tritium.
2) The tritium release from the Li_2TiO_3 pebble bed was evaluated for the open and closed conditions of the Hf window. The time constants of the changes in the irradiation temperature and the tritium release were about 100 and 18,000 s, respectively.
3) The tritium release rate increased cycle by cycle. After about 20 pulses, the average tritium release rate was almost constant, and the normalized tritium release rate $(R/G)_{av}$ approached unity. It is found from this test that the number of pulse operation cycles to obtain unity for $(R/G)_{av}$, depends on the pulse operation condition and irradiation temperature.
4) When the temperature at the outside edge of the Li_2TiO_3 pebble bed exceeded 100°C, the tritium release from Li_2TiO_3 pebble bed began to be observed. The normalized tritium release

rate (R/G) reached unity when the temperature at the outside edge of the Li_2TiO_3 pebble bed was kept above 300°C.

5) The change in the sweep-gas flow rate was followed by positive tritium release peaks for increase in the sweep gas flow rate, and by negative release peaks for decrease in the sweep gas flow rate. On the other hand, the sweep gas flow rate had not effect on tritium release from the Li_2TiO_3 pebble bed in the steady state for the range of the sweep gas flow rate from 100 to 900cm^3/min (superficial gas velocity in the Li_2TiO_3 pebble bed : 0.53 – 4.8 cm/s).
6) When the hydrogen partial pressure was higher than 1 Pa, the total tritium release rate was constant. However, the total tritium release rate decreased with decreasing the hydrogen partial pressure when the hydrogen partial pressure was lower than 1 Pa. These results of the hydrogen content changing tests show that the hydrogen partial pressure in the sweep gas had an effect on tritium release from the Li_2TiO_3 pebble bed.

From these results of the in-situ experiments, the Li_2TiO_3 pebbles fabricated by the wet process appear to be promising as a candidate tritium breeder for the blanket of ITER.

7. Future Plan

It is necessary to investigate the following properties by in-situ irradiation tests with a Li_2TiO_3 pebble bed in future.

- Development of tritium release model for a Li_2TiO_3 pebble bed.
- Effect of the irradiation temperature on tritium release from a Li_2TiO_3 pebble bed below 300°C.
- Effect of different pulsed operation modes on tritium release from a Li_2TiO_3 pebble bed.
- Effect of Li burn-up (up to about 5% Li-burnup) on tritium release from a Li_2TiO_3 pebble bed.

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Table 1 Impurities in Li_2TiO_3 powder.

Element	Measured values (wt ppm)	Detection limit (wt ppm)
B	< 5	< 5
Na	16	< 5
Mg	4	< 2
Al	< 10	< 10
Si	27	< 10
K	< 5	< 5
Ca	< 2	< 2
Cr	7	< 2
Mn	2	< 2
Fe	8	< 2
Co	< 0.5	< 0.5
Ni	< 5	< 5
Cu	< 2	< 2
Zr	< 2	< 2
U	< 0.1	< 0.1

Table 2 Characteristics of two kinds of Li_2TiO_3 pebbles.

properties \ Pebbles	Large Pebble	Small Pebble
Material	Li_2TiO_3	Li_2TiO_3
Fabrication Method	Wet process with dehydration reaction	Wet process with substitution reaction
Diameter	ϕ1.93 mm av. (ϕ1.7 - 2.36 mm)	ϕ0.29 mm av. (ϕ0.25 - 0.3 mm)
Density	2.86 g/cm^3 (83.4%T.D.)	2.82 g/cm^3 (82.2%T.D.)
Sphericity	1.07	1.11
Grain size	< 5 μm	< 5 μm
Collapse Load	73.4 N	4.57 N
Impurity	Ca;<2, Na;82, Al;9, Si;14 (wtppm)	Ca;18, Na;39, Al;23, Si;73 (wtppm)

Table 3 Impurity contents of two kinds of Li_2TiO_3 pebbles.

Elements	Measured values (wt ppm)		Detection Limit (wt ppm)
	Large Pebble	Small Pebble	
Ca	< 2	18	< 2
Na	82	39	< 2
K	< 2	2	< 2
Mg	< 2	6	< 2
B	< 5	< 5	< 5
Co	< 0.5	< 0.5	< 0.5
Al	9	23	< 10
Si	14	73	< 10
Zr	< 2	< 2	< 2
Fe	< 2	25	< 2
Zn	2	22	< 2
Mn	< 2	< 2	< 2
C	77	20	< 1
Cl	6	2	< 1
Cu	< 2	2	< 2
U	< 0.1	< 0.1	< 0.1

Table 4 Calculation parameters for irradiation temperature in pebble bed.

(1) Cooling Condition
- Temperature of Cooling Water : 50°C
- Heat Transfer Coefficient : 2.33W/cm°C
(Calculated value from the cooling condition of JMTR)

(2) Property Values
- Lithium Titanate (Li_2TiO_3)
  - a) Pebble Bed Density : 2.28g/cm^3
  - b) Theoretical Density : 3.43g/cm^3
  - c) Sintering Density : 83%T.D.
  - d) Packing Fraction : 80%
    - Primary pebble (large) : 63%
    - Secondary pebble (small) : 17%
  - c) Effective Thermal Conductivity (W/cm°C) (Evaluation from refs.[19] and [20])

$$\lambda=1.24\times10^{-2}+2.33\times10^{-7}\cdot T-3.35\times10^{-9}\cdot T^2+7.65\times10^{-12}\cdot T^3$$

- Hafnium (Hf) (ref. [24])
  - a) Density=13.36g/cm^3
  - b) Thermal Conductivity (W/cm°C) (Evaluation from ref.[24])

$$\lambda=0.230\times10^{0}-7.000\times10^{-5}\cdot T+5.400\times10^{-8}\cdot T^2$$

  - c) Thermal Expansion=5.9×10^{-6} (1/K)

Table 5　Calculation results of in-pile mockup by MCNP code and GENGTC code.

Items \ Condition	Condition of Hf Window	
	Open	Close
Fast Neutron Fluence ($n/m^2/s$)	5.4×10^{15}	4.4×10^{15}
Thermal Neutron Fluence ($n/m^2/s$)	1.9×10^{16}	2.7×10^{15}
Total Neutron Fluence ($n/m^2/s$)	8.2×10^{16}	5.2×10^{16}
α-heat (kW/kg)	0.75	0.11
γ-heat (kW/kg)	0.17	0.14
Tritium Generation (Bq/d)	3.3×10^{10}	4.8×10^{9}
Center Temperature of Pebble Bed (°C)	356	275
Temperature at Edge of Pebble Bed (°C)	315	264

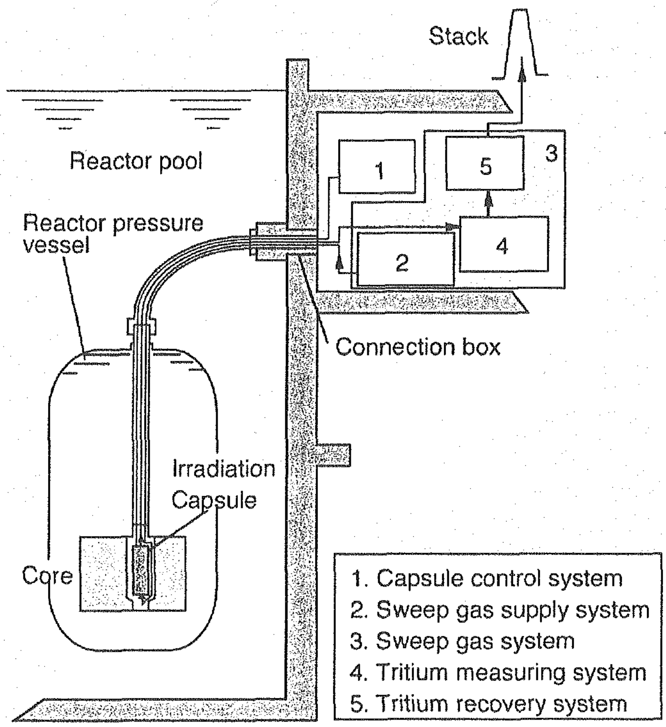


Fig.1 JMTR irradiation facility for in-situ irradiation test (reprinted from "K. Tsuchiya, et al., J. Nucl. Sci. Technol. 38 (2001) 996" [13]).

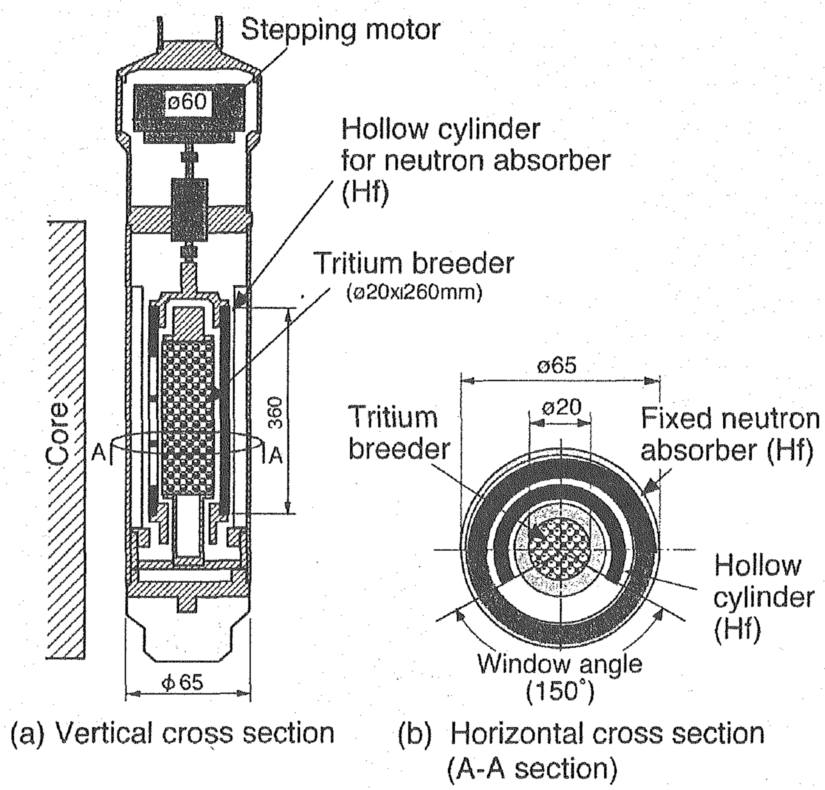


Fig.2 Conceptual design of pulse-operation simulating mockup.

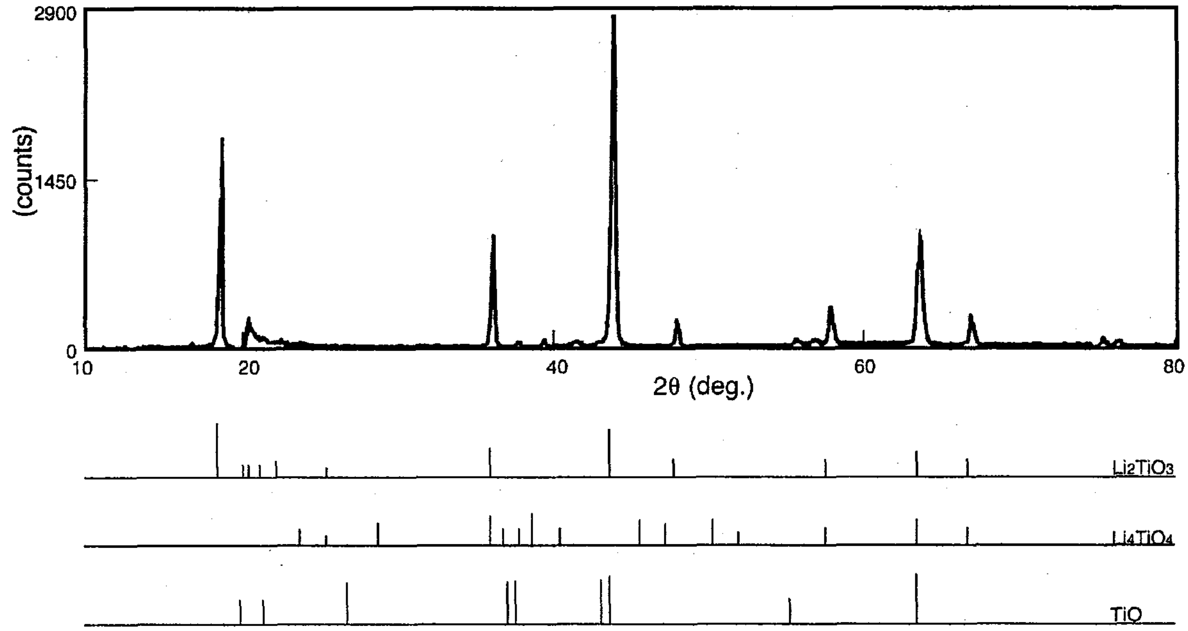
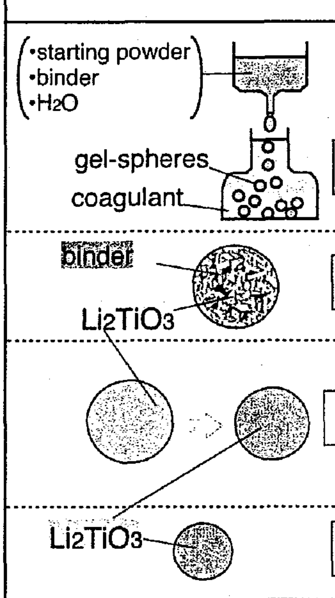


Fig.3 X-ray diffraction patterns of Li_2TiO_3 powder.

Fabrication Process	Wet process with dehydration reaction	Wet process with substitution reaction
Mixing	binder : polyvinyl alcohol (PVA) *1 coagulant : acetone	binder : ammonium alginate*1 coagulant : zinc chloride*1
Dropping	coagulant Temp. : <-20°C	coagulant Temp. : RT
Drying	drying time : ~24 h	drying time : ~0.5 h
Calcination	in air ~650°CX6h	reduction (H_2 gas) ~1000°CX4h
Sintering	D_{min}*2=0.5mm	D_{min}*2=~0.2mm



*1 : candidate material, *2 : minimum diameter of pebbles with high sphericity

Fig.4 Outline of pebble fabrication by wet process.

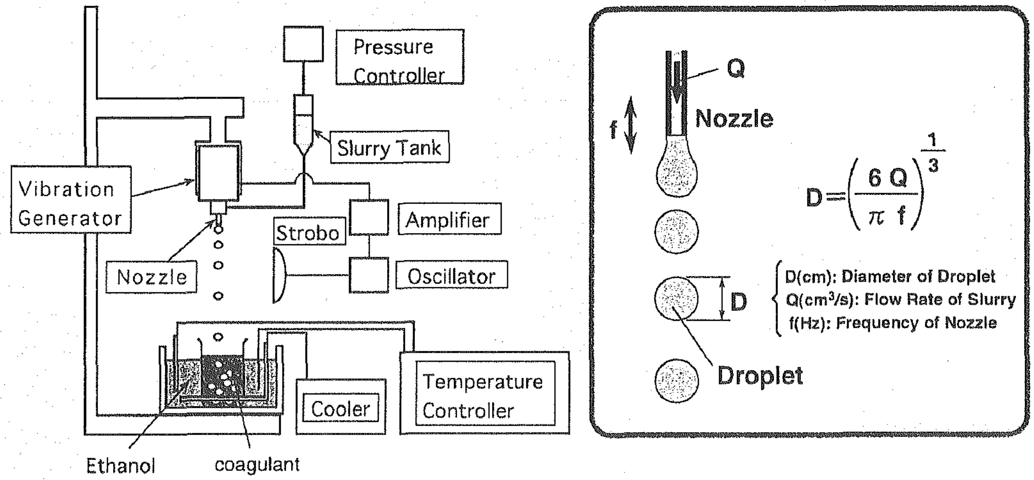


Fig.5 Schematic drawing of vibratory dropping system.

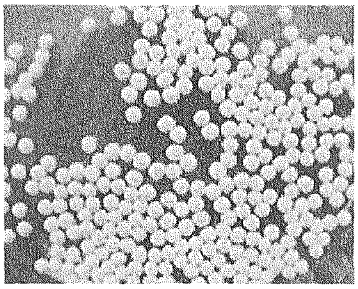
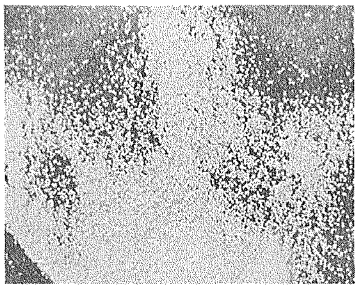
	Li_2TiO_3 Pebbles	
	Large pebbles (ϕ 1.93 mm av.)	Small pebbles (ϕ 0.29 mm av.)
Photograph		

Fig.6 Photographs of two kinds of Li_2TiO_3 pebbles.

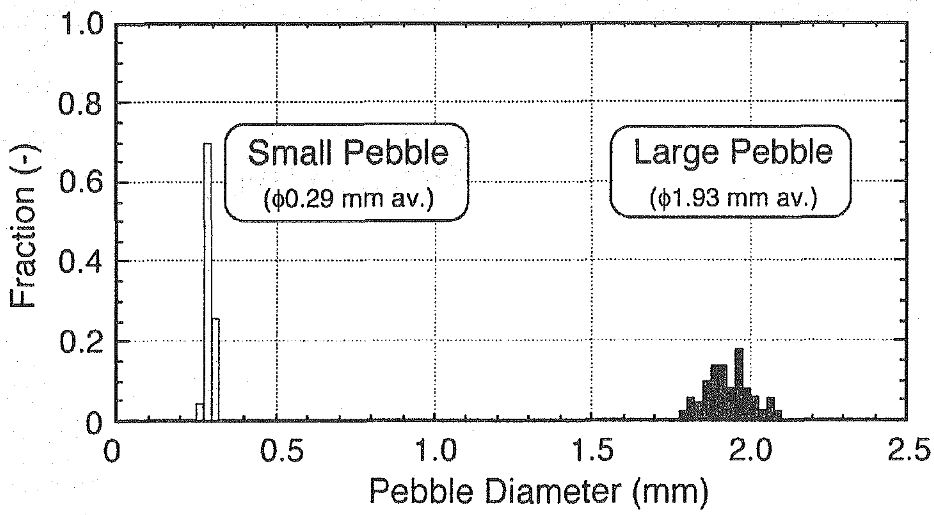
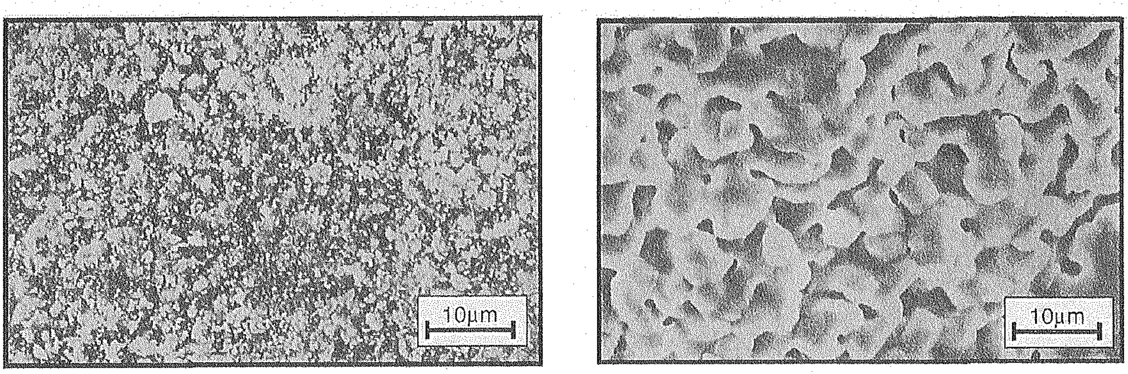


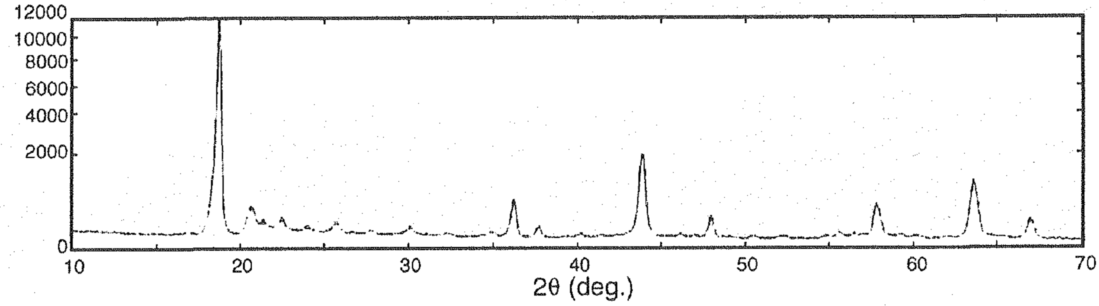
Fig.7 Distribution on diameter of two kinds of Li_2TiO_3 pebbles fabricated by wet process.



a) Large Pebble (ϕ1.93 mm av.)　　b) Small Pebble (ϕ0.29 mm av.)

Fig.8　SEM photograph of cross section of Li_2TiO_3 pebbles.

a) Large Pebble (ϕ1.93 mm av.)



b) Small Pebble (ϕ0.29 mm av.)

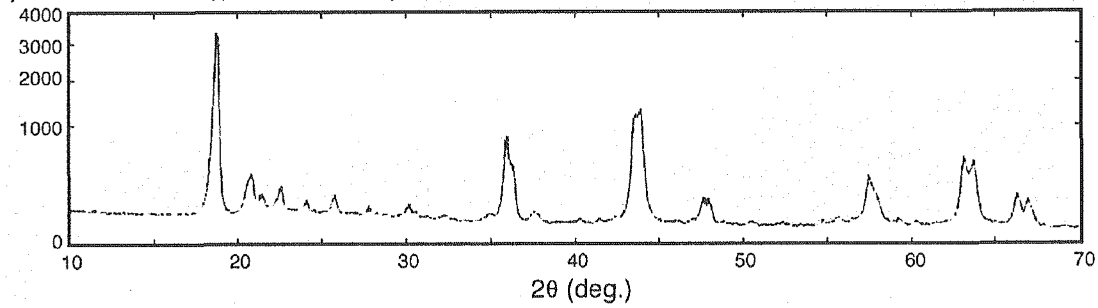


Fig.9　X-ray diffraction patterns of two kinds of Li_2TiO_3 pebbles.

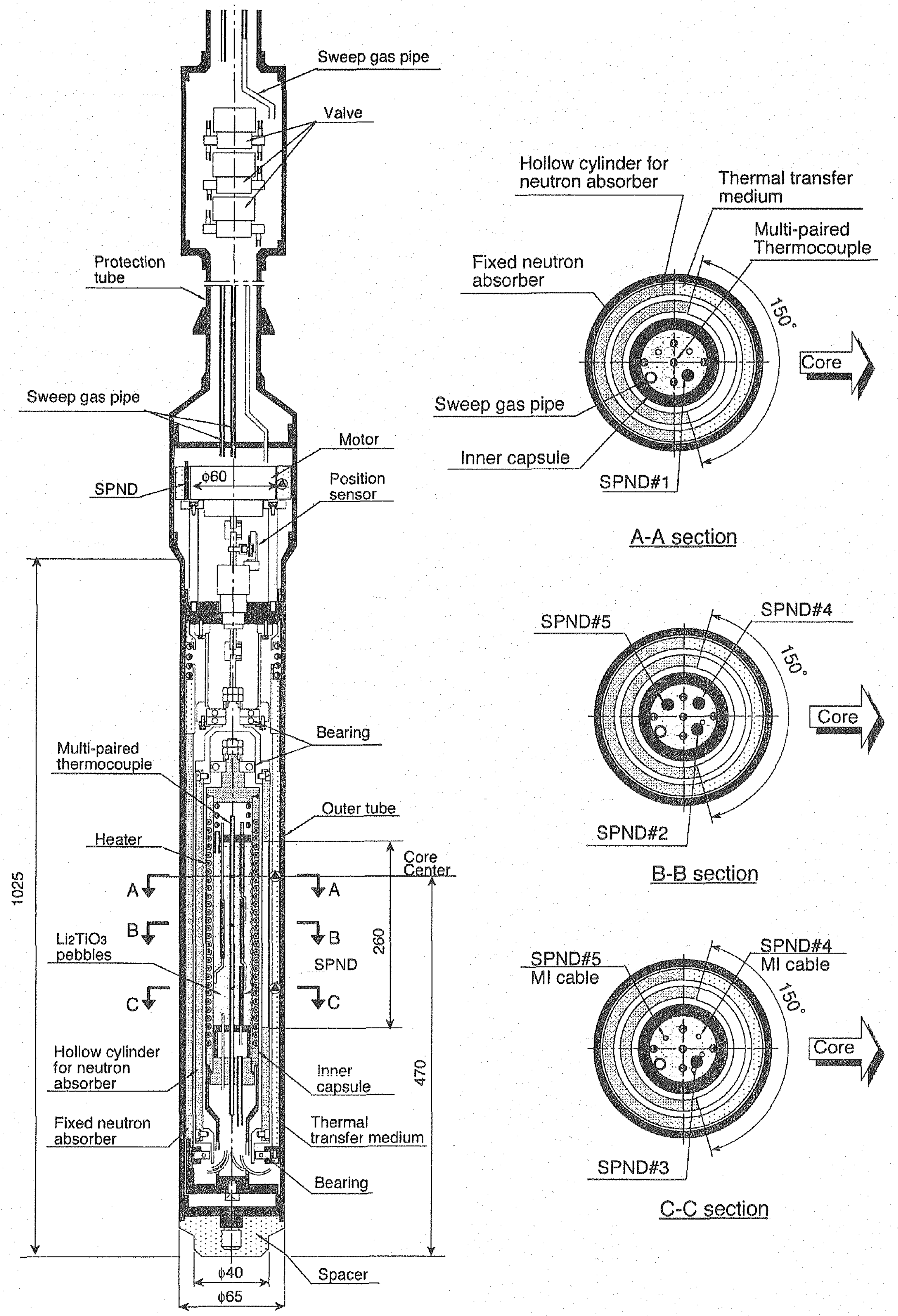


Fig.10 Cross section of pulse-operation simulating mockup.

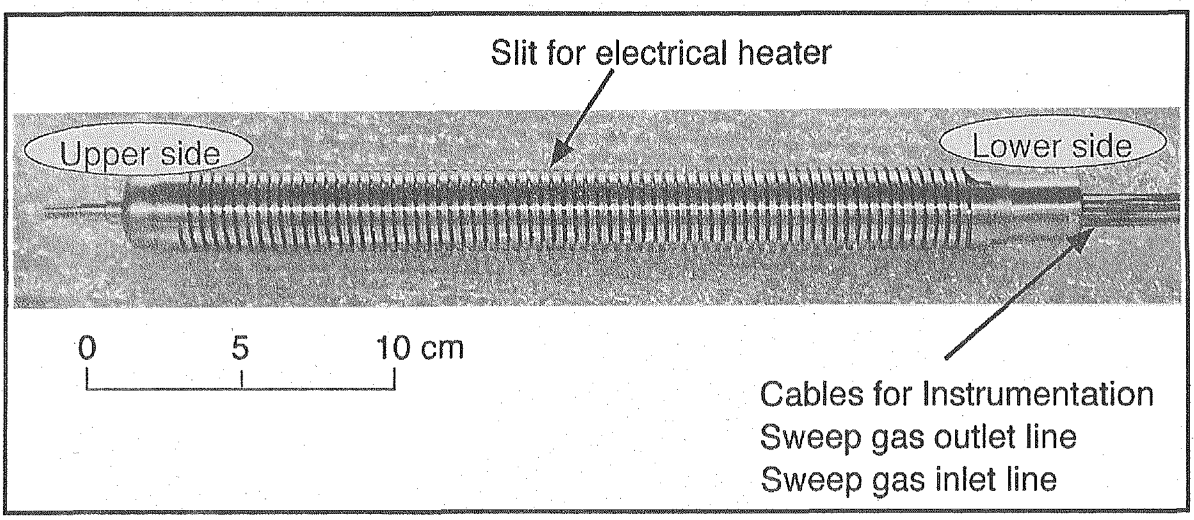


Fig.11 Photograph of inner capsule.

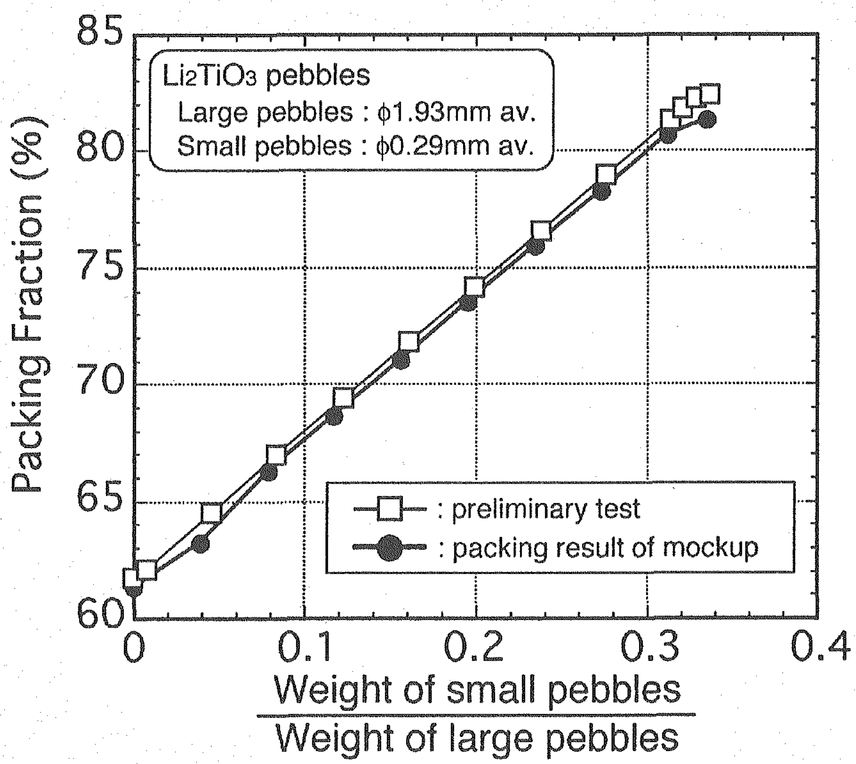


Fig.12 Results of packing fraction measurements of Li_2TiO_3 pebbles.

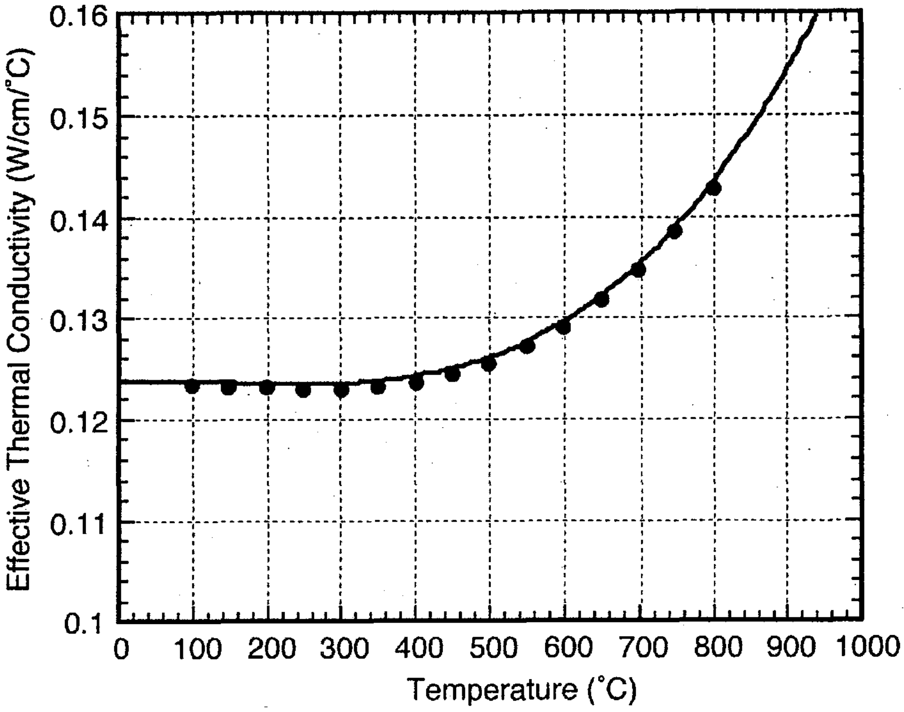


Fig.13 Calculated effective thermal conductivity of binary Li_2TiO_3 pebble bed. (see section 3.1 in the text).

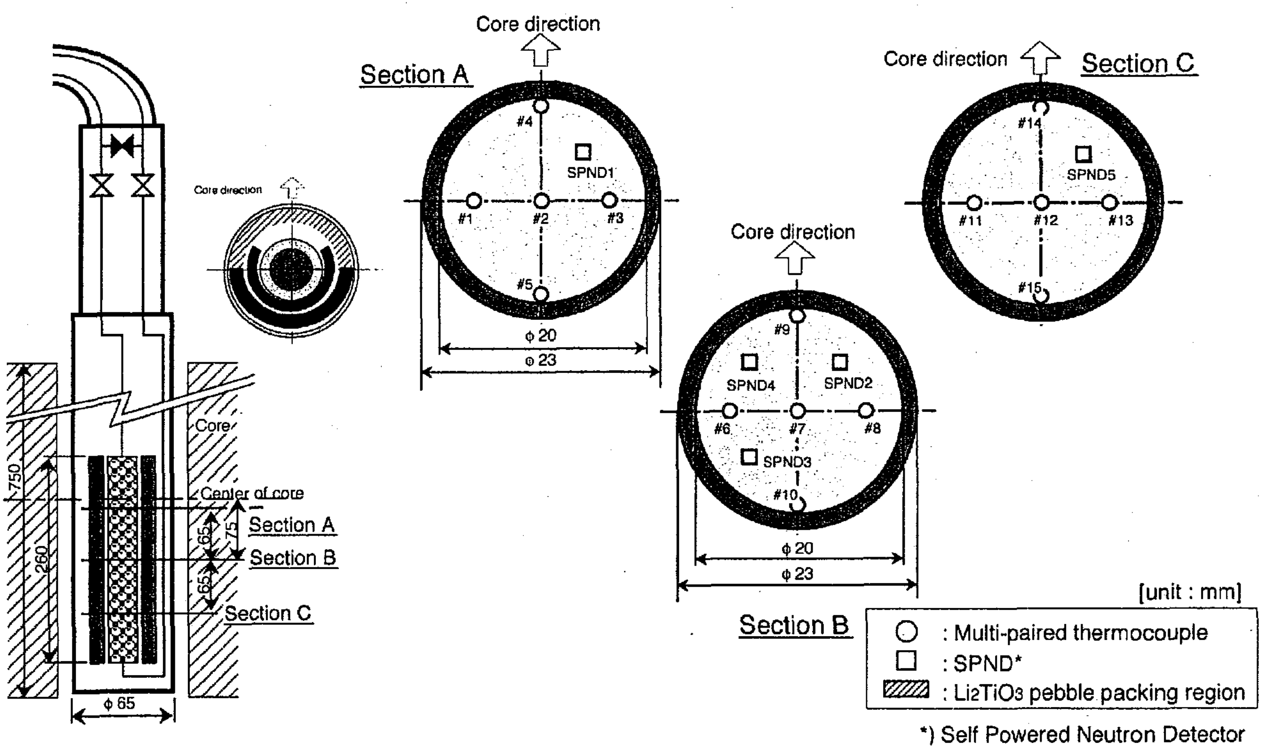
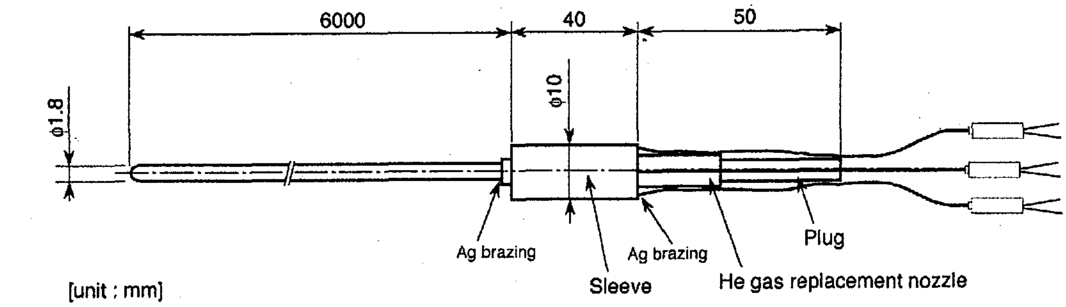
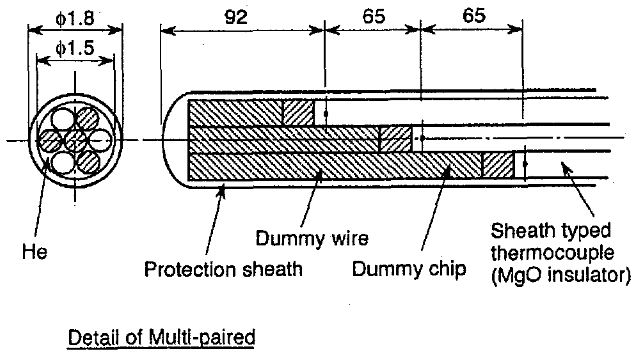


Fig.14 Arrangement of thermocouples and self-powered neutron detectors (SPND).

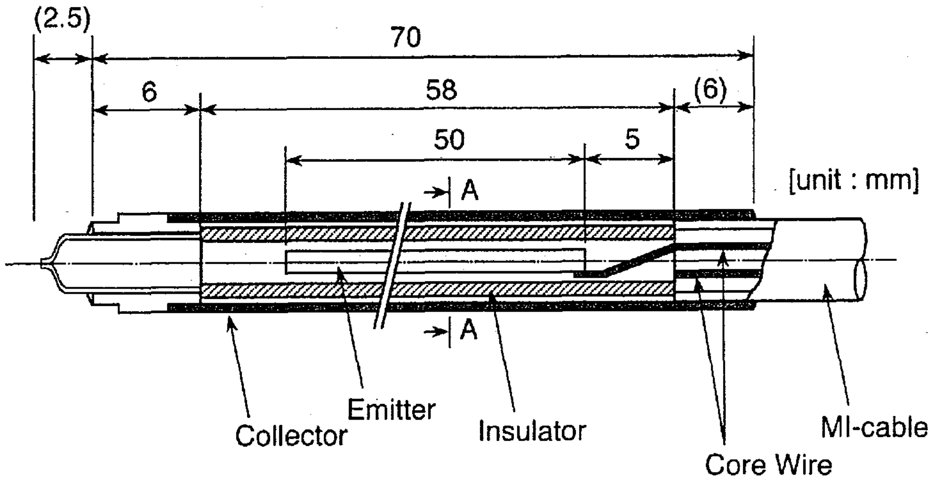




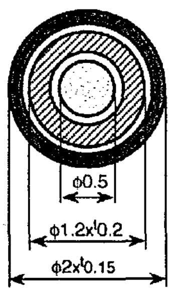
Specification of Multi-paired Thermocouple

Items	Material	Remark
Sheath typed thermocouple	SS316L	Sheath O.D. : φ0.5 Type : K (CA)
Dummy wire	SS304	
Protection sheath	Inconel600	
Sleeve	SS304	
He gas replacement nozzle	SS304	
Plug	SS304	

Fig.15 Structure and specification of multi-paired thermocouples.



A-A Cross Section



Specification of SPND

Part of measurement			MI-cable		
Emitter	Collector	Insulator	Core wire (twin)	Insulator	Sheath
Pt-Rh	Inconel 600	Al_2O_3 (99.5wt.%)	Inconel 600	Al_2O_3 (99.6wt.%)	Inconel 600 (φ1.5)

Fig.16 Structure and specification of self-powered neutron detector (SPND).

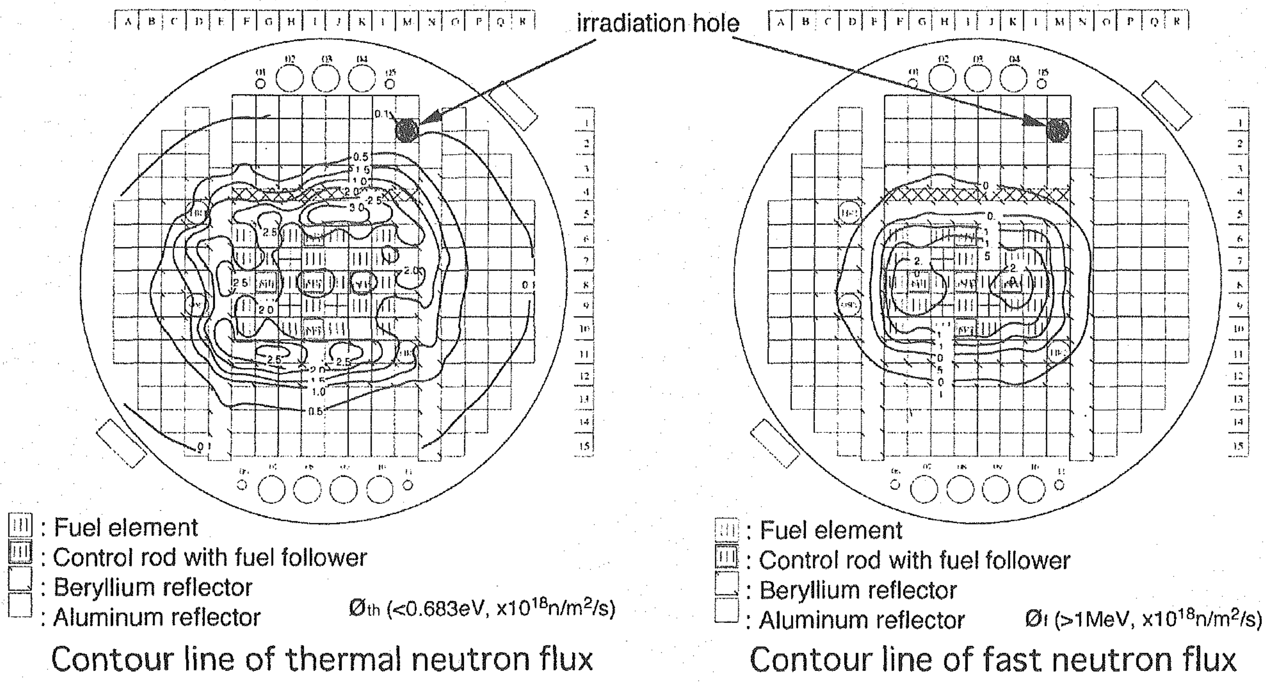


Fig.17 Irradiation hole for irradiation capsule.

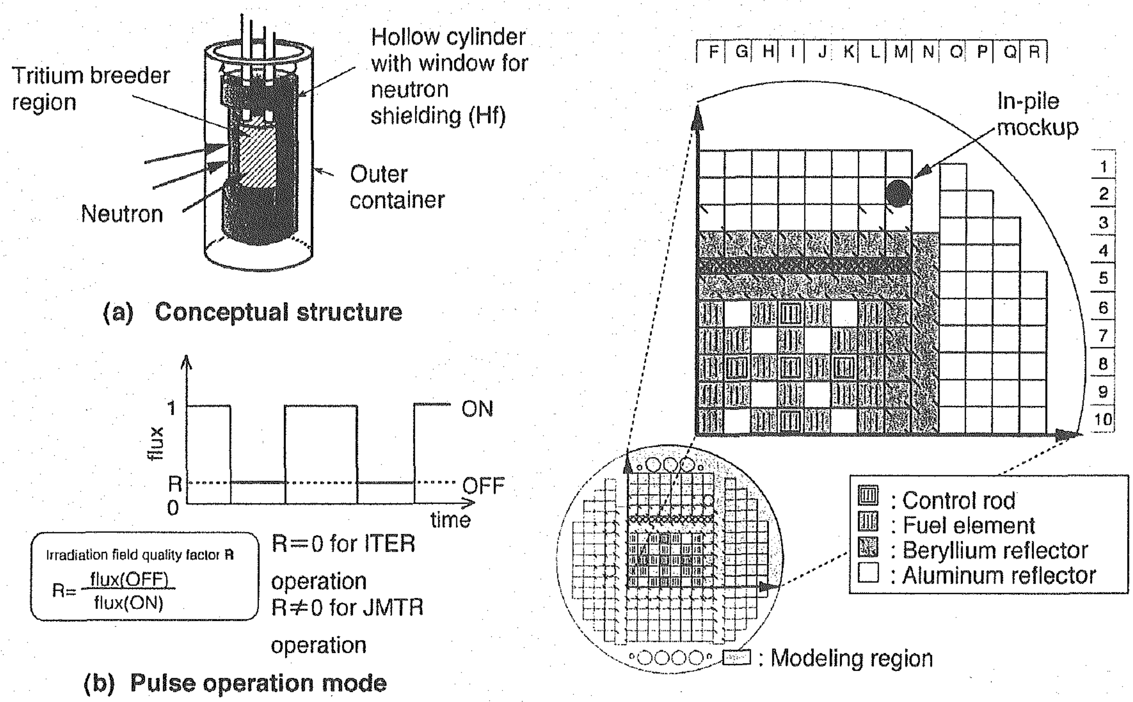


Fig.18 Conceptual structure of the in-pile mockup and calculation model on JMTR core.

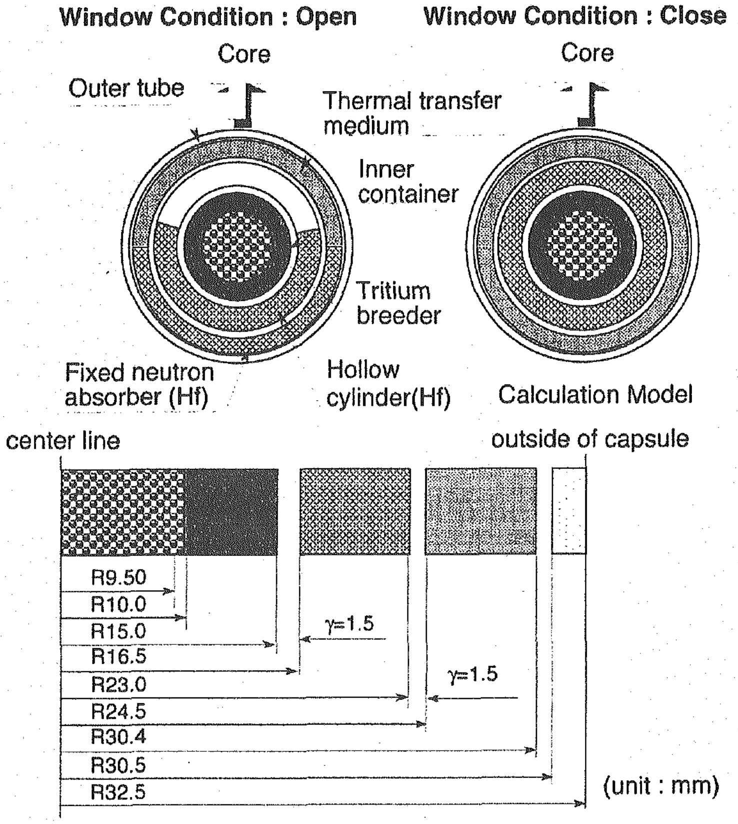


Fig.19 Calculation model of irradiation temperature in pebble bed.

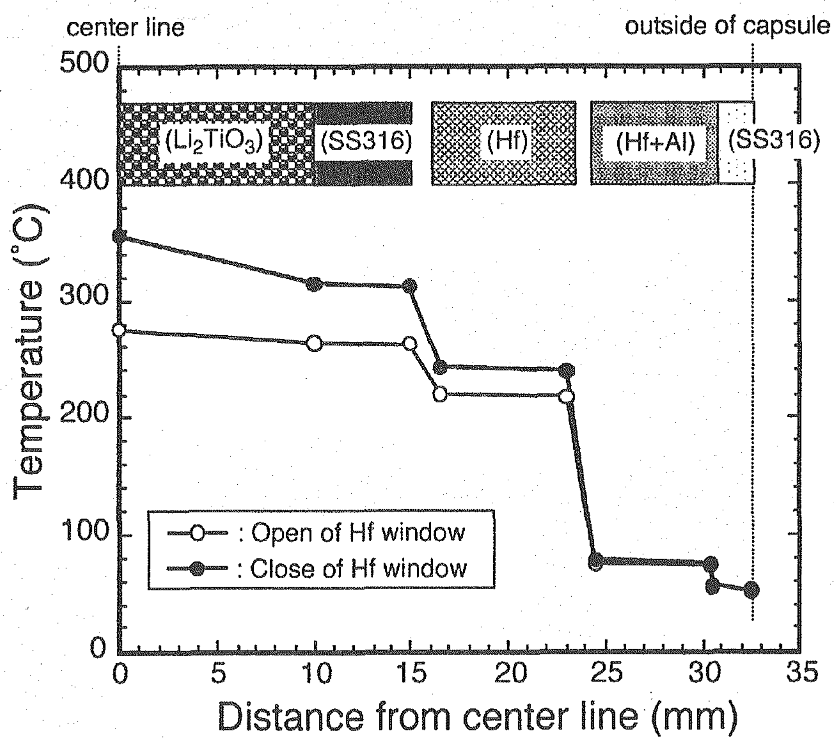
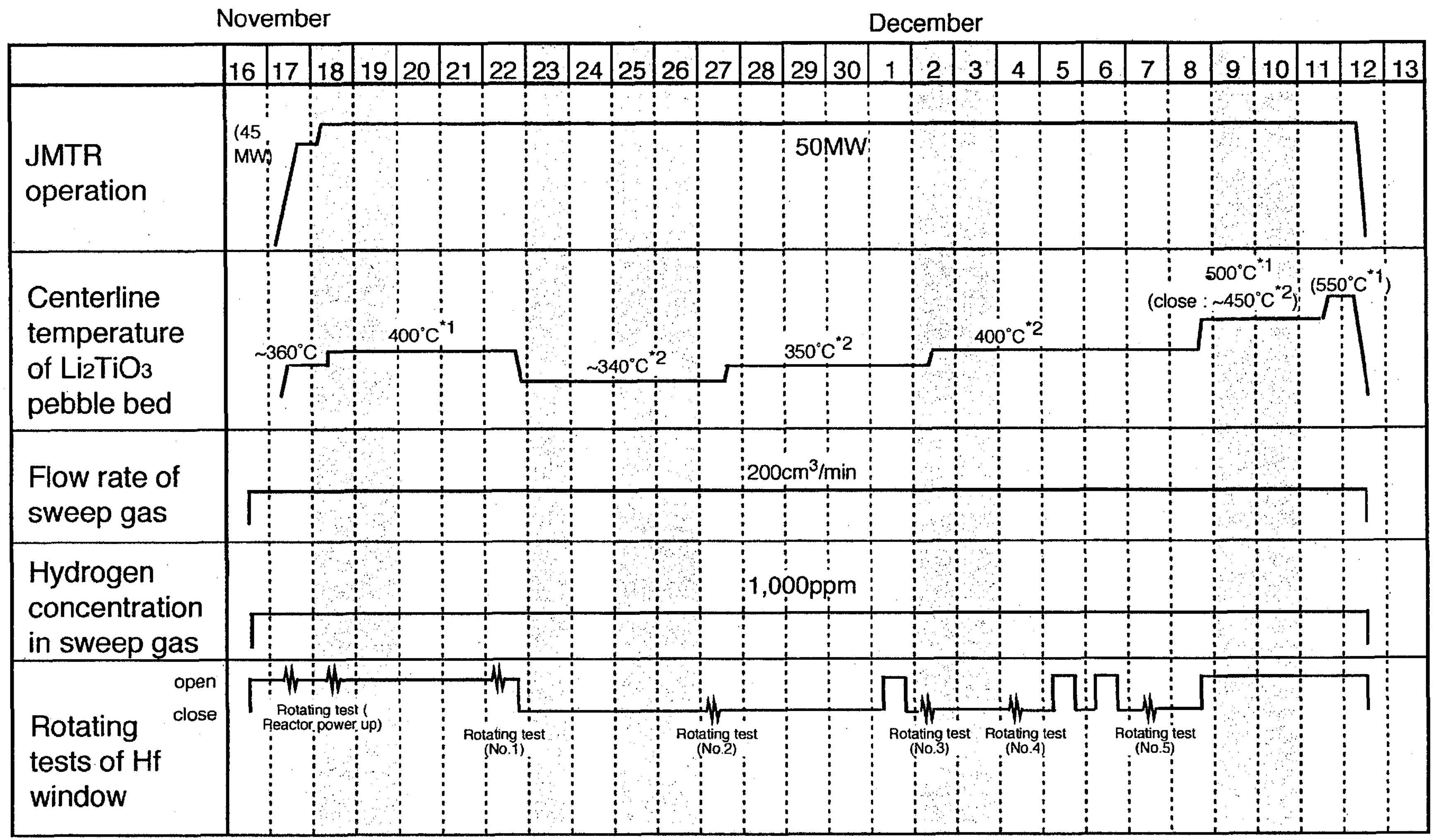


Fig.20 Result of nuclear and thermal calculation of irradiation capsule.



*1 : Center temperature of Li_2TiO_3 pebble bed when Hf window was opened.
*2 : Center temperature of Li_2TiO_3 pebble bed when Hf window was closed.

Fig.21 Outline of experimental conditions in the first irradiation test.

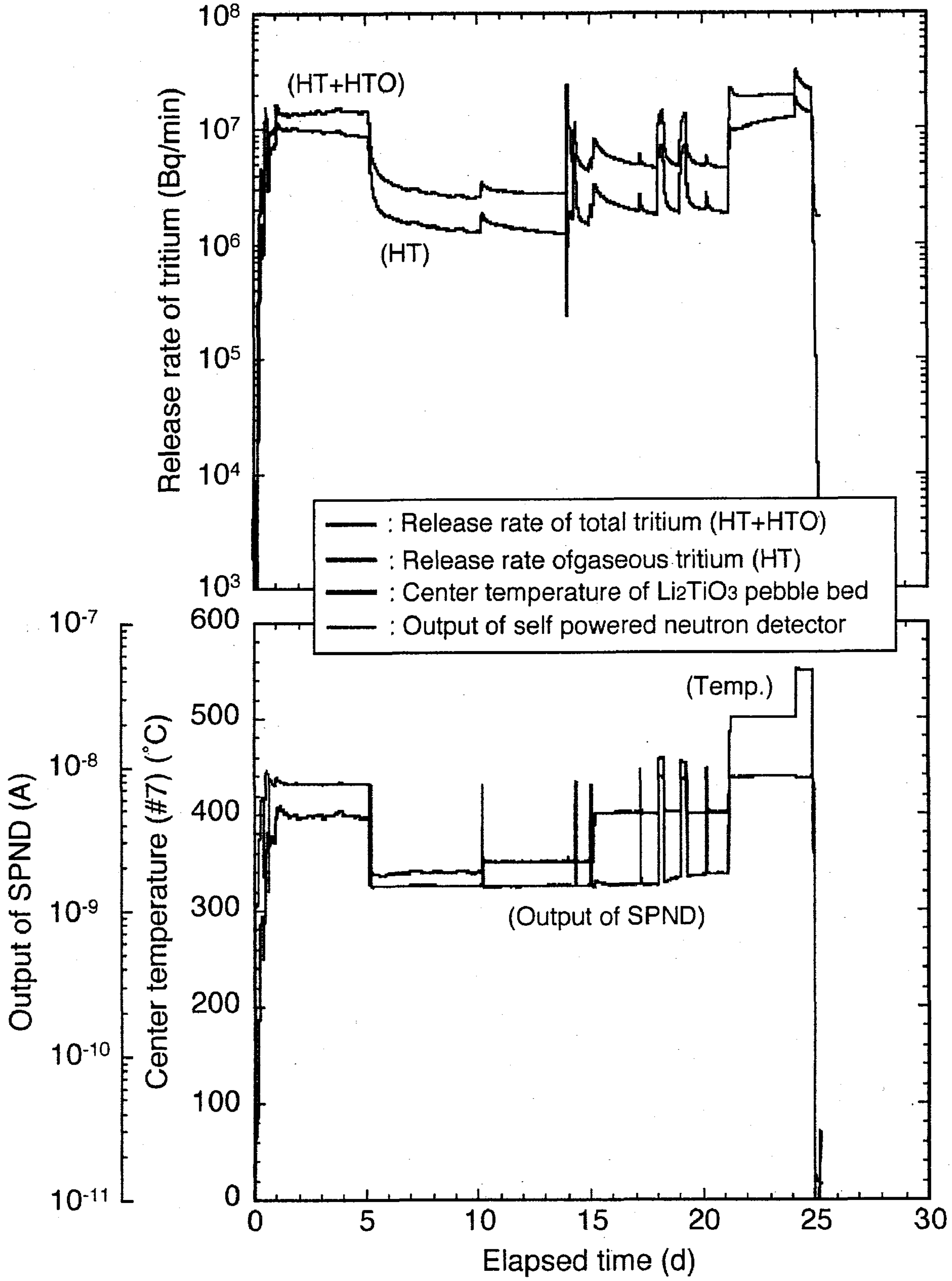


Fig.22 Result of the first irradiation test in the 136th cycle of JMTR.

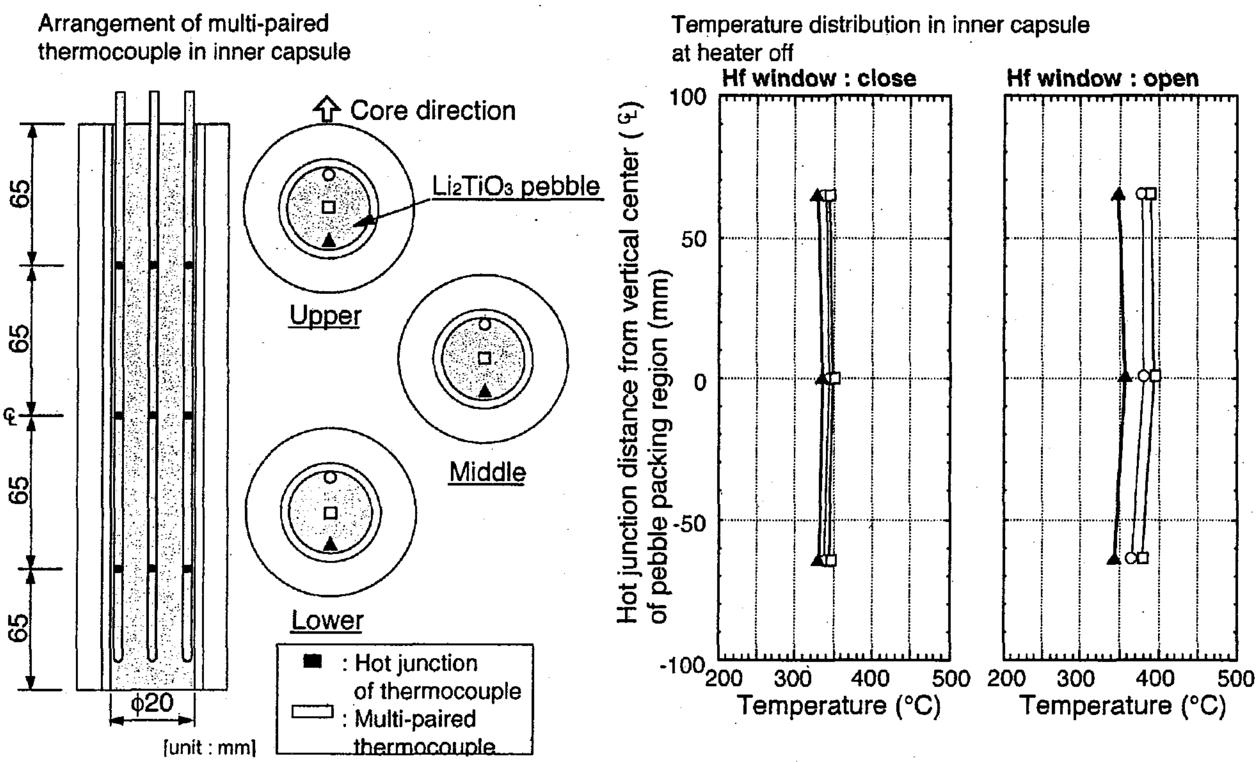


Fig.23 Vertical temperature distribution in the first irradiation test.

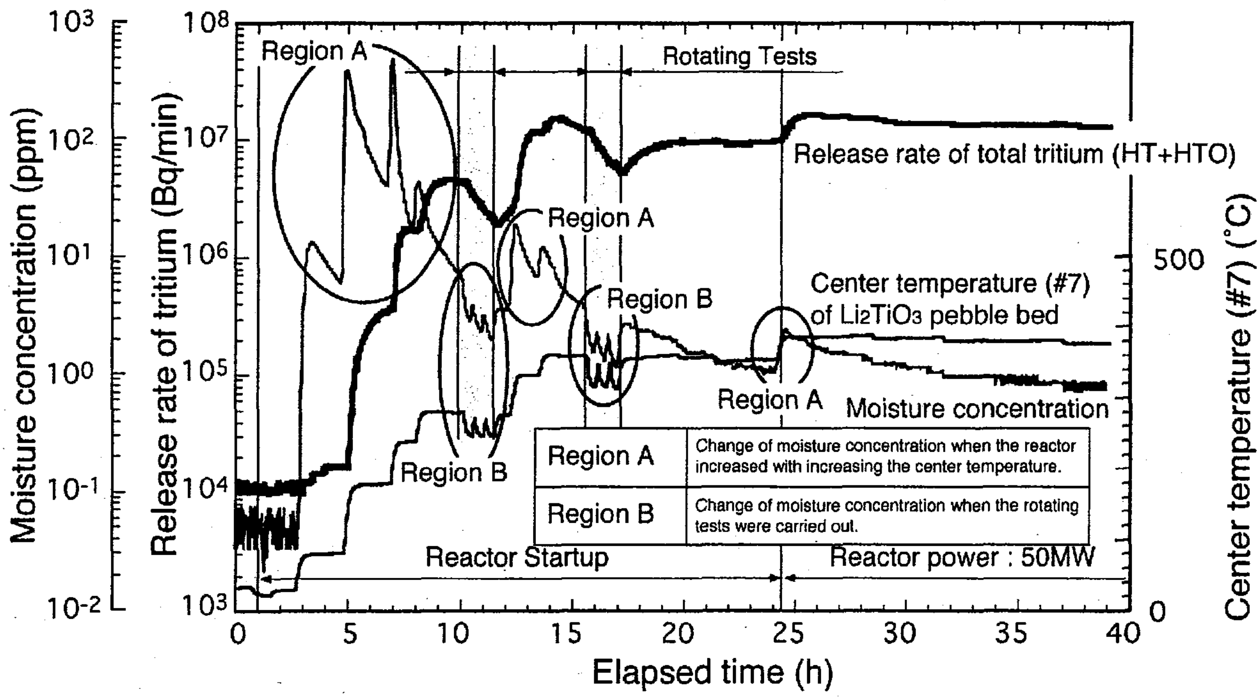


Fig.24 Tritium release at reactor power-up in the first irradiation test.

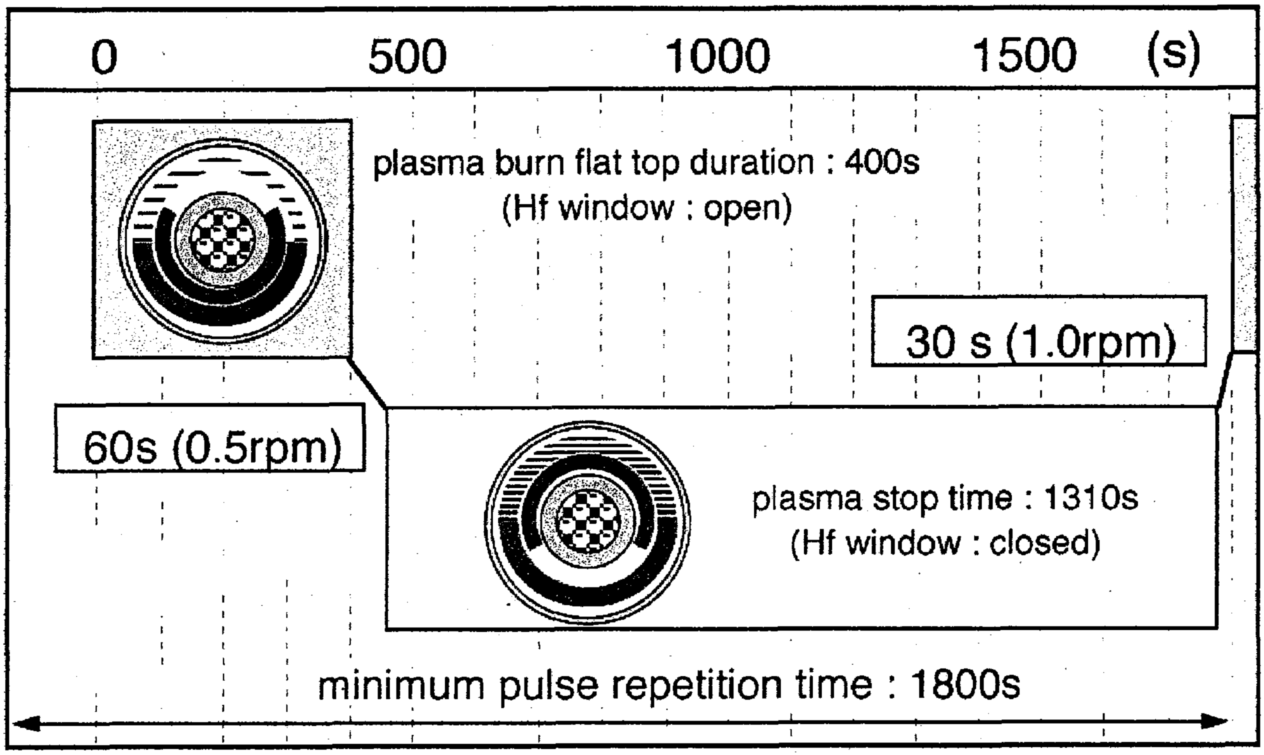


Fig.25 Rotating condition in nominal pulsed operation of ITER.

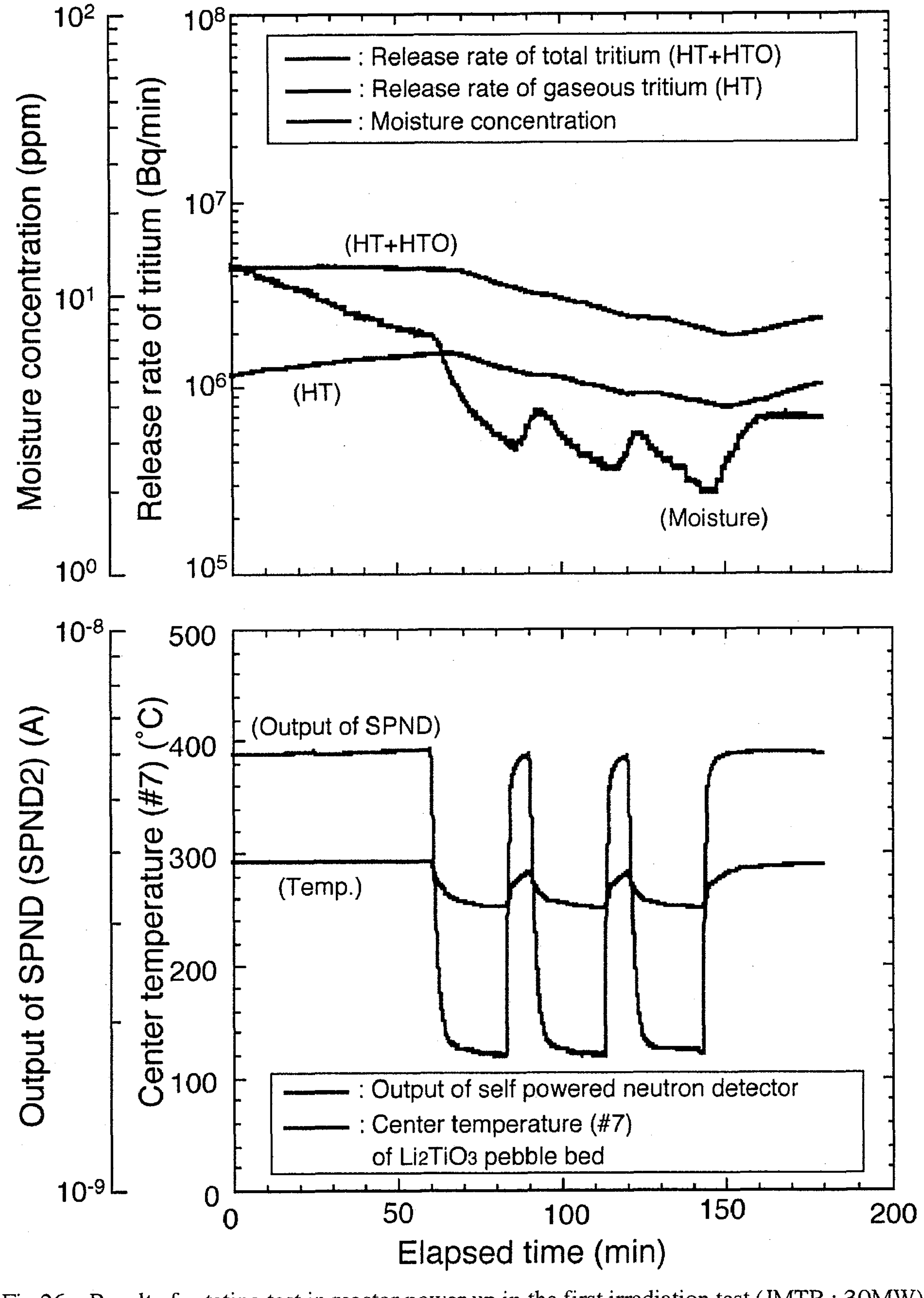


Fig.26 Result of rotating test in reactor power up in the first irradiation test (JMTR : 30MW).

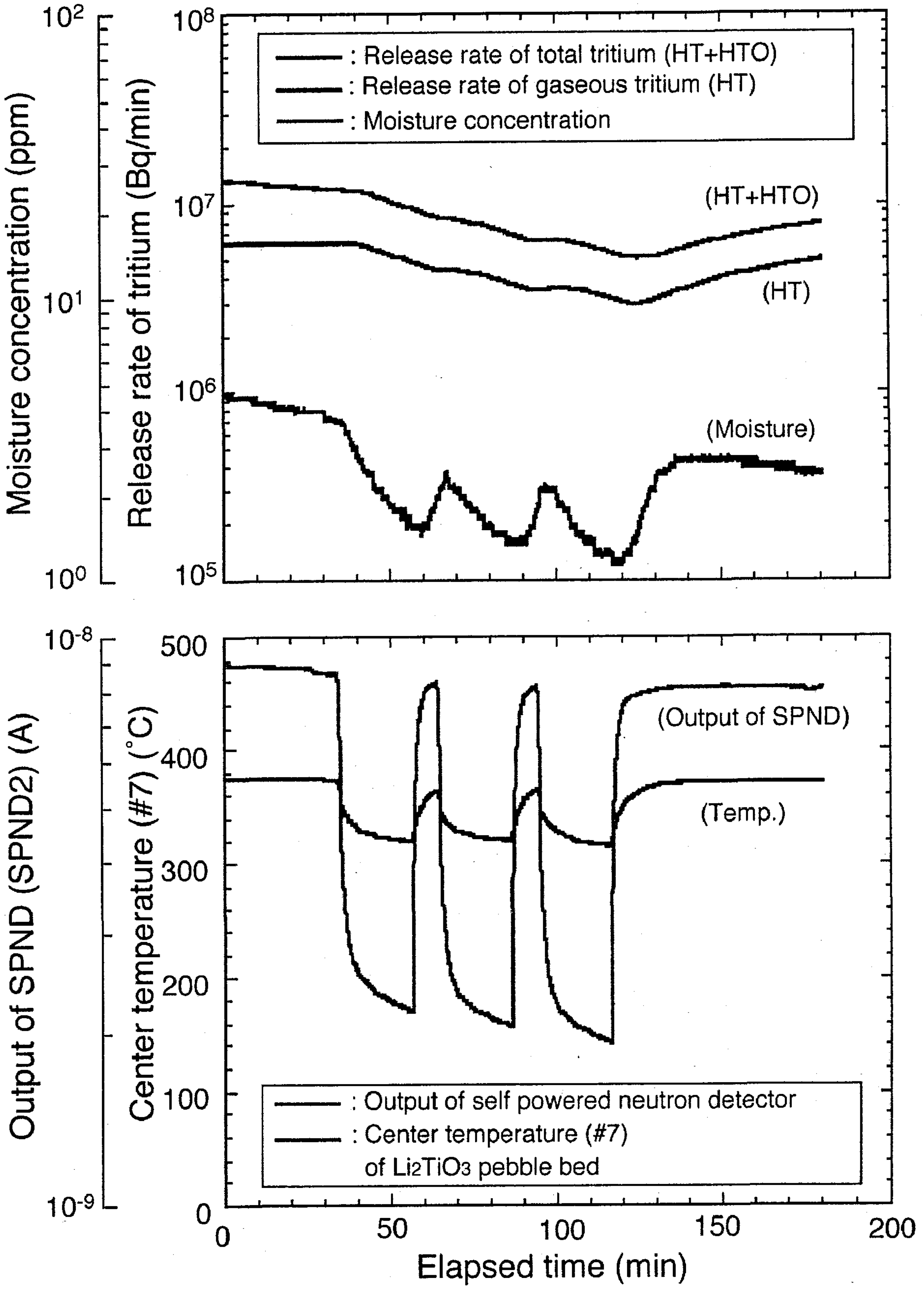


Fig.27 Result of rotating test in reactor power up in the first irradiation test (JMTR : 45MW).

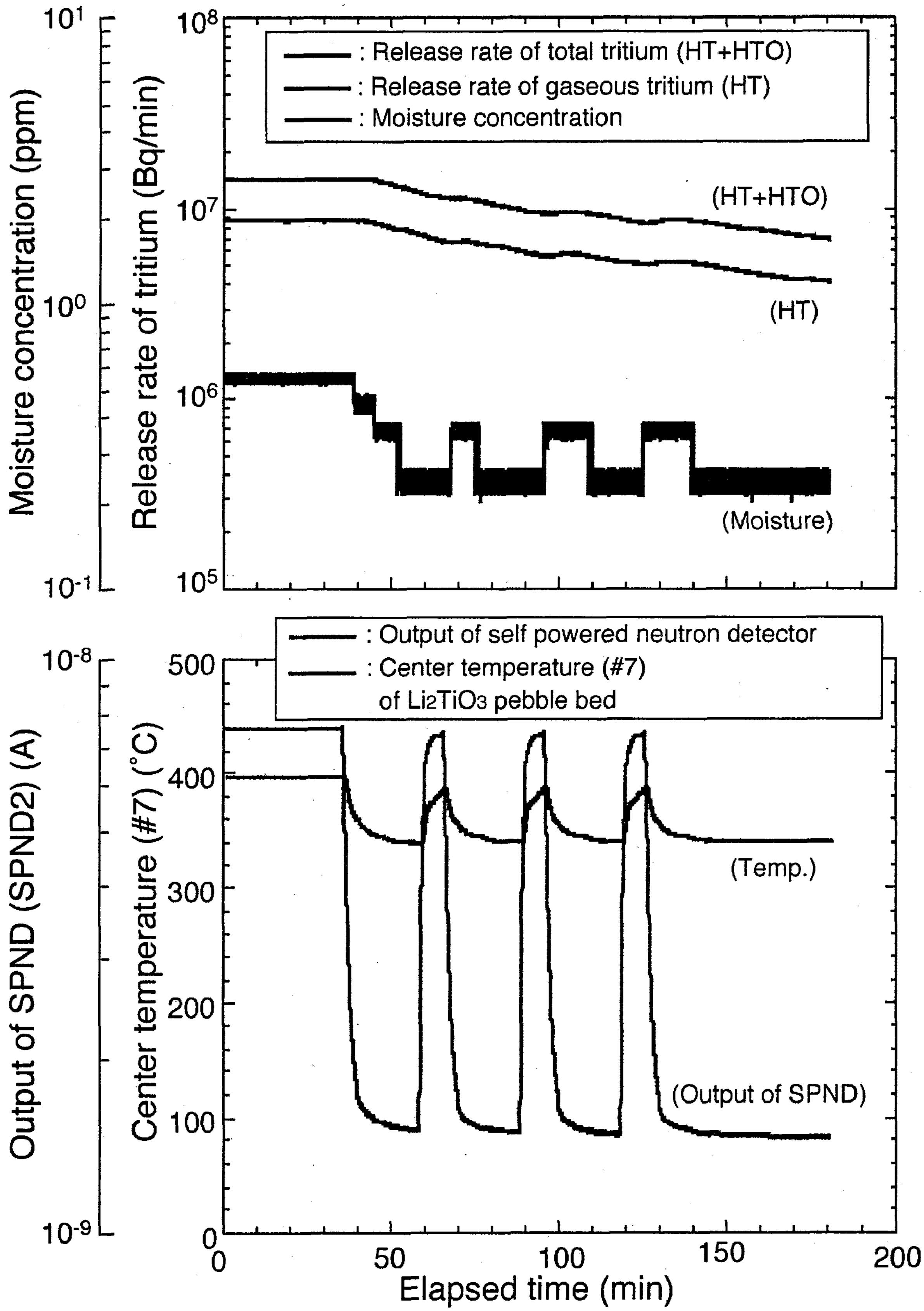
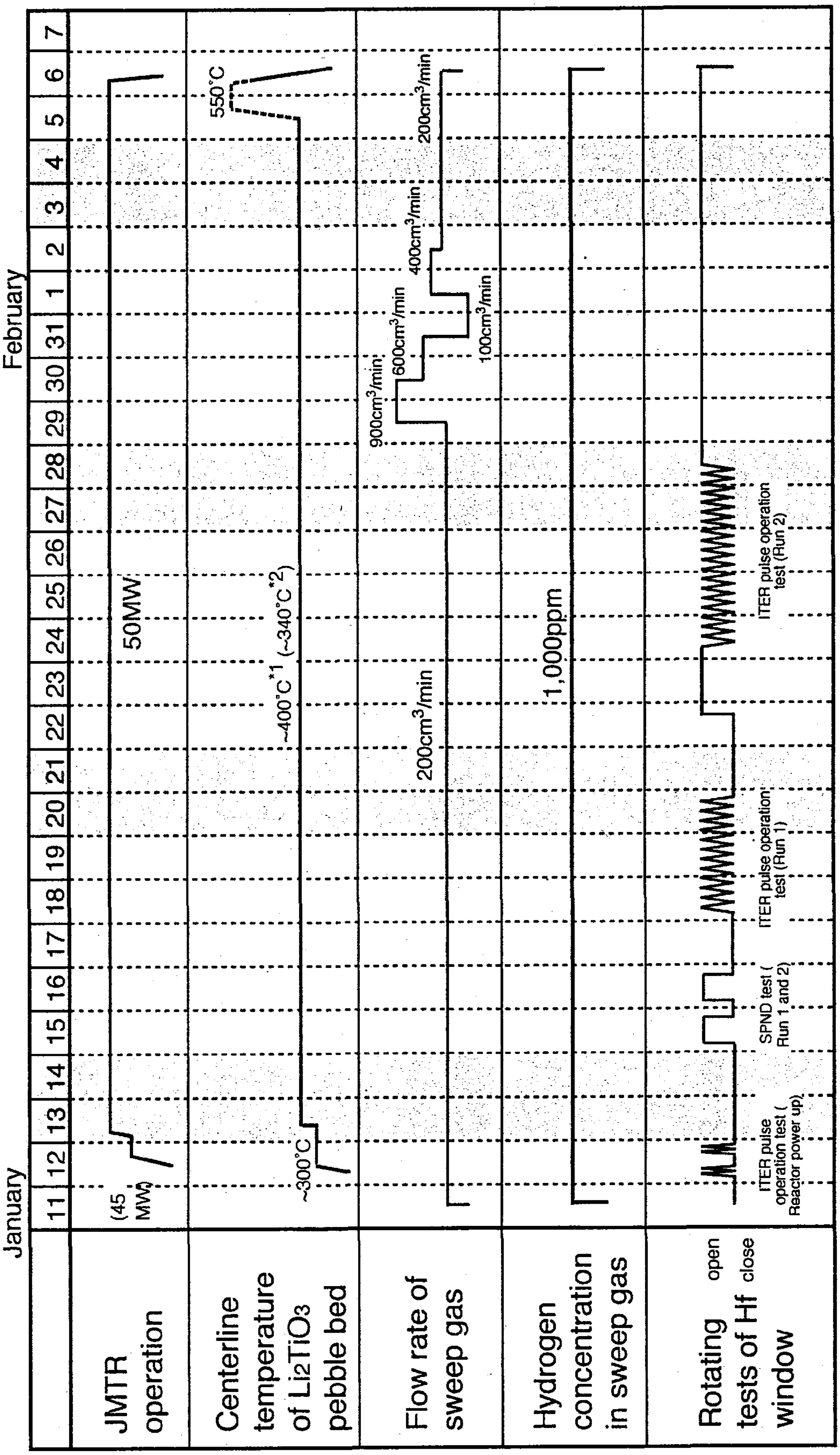


Fig.28 Result of rotating test in reactor power up in the first irradiation test (JMTR : 50MW).



*1 : Center temperature of Li_2TiO_3 pebble bed when Hf window was opened.
*2 : Center temperature of Li_2TiO_3 pebble bed when Hf window was closed.

Fig.29 Outline of experimental conditions in the second irradiation test.

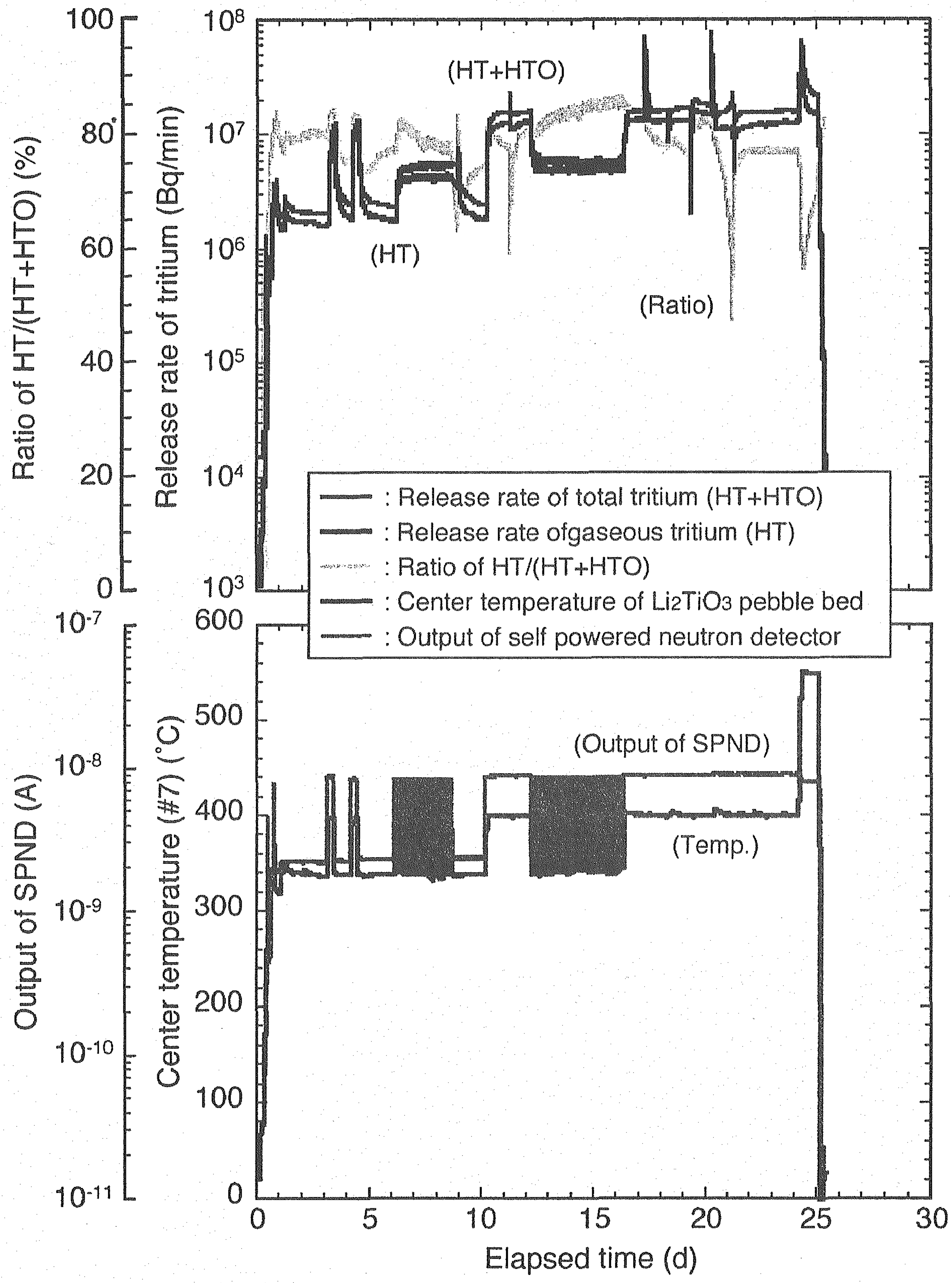


Fig.30 Result of the second irradiation test in the 137th cycle of JMTR.

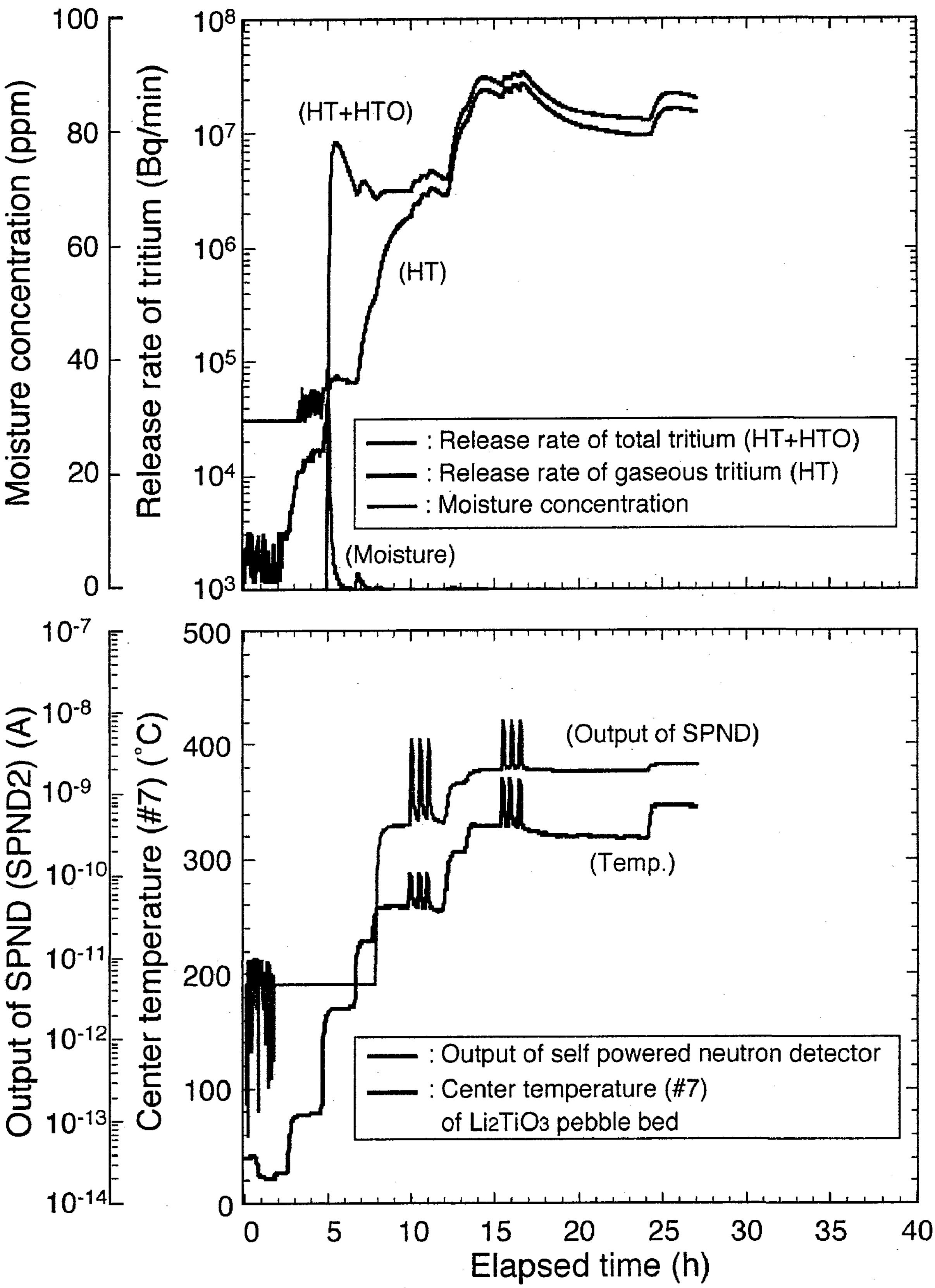


Fig.31 Result of tritium release at the reactor startup in the second irradiation test.

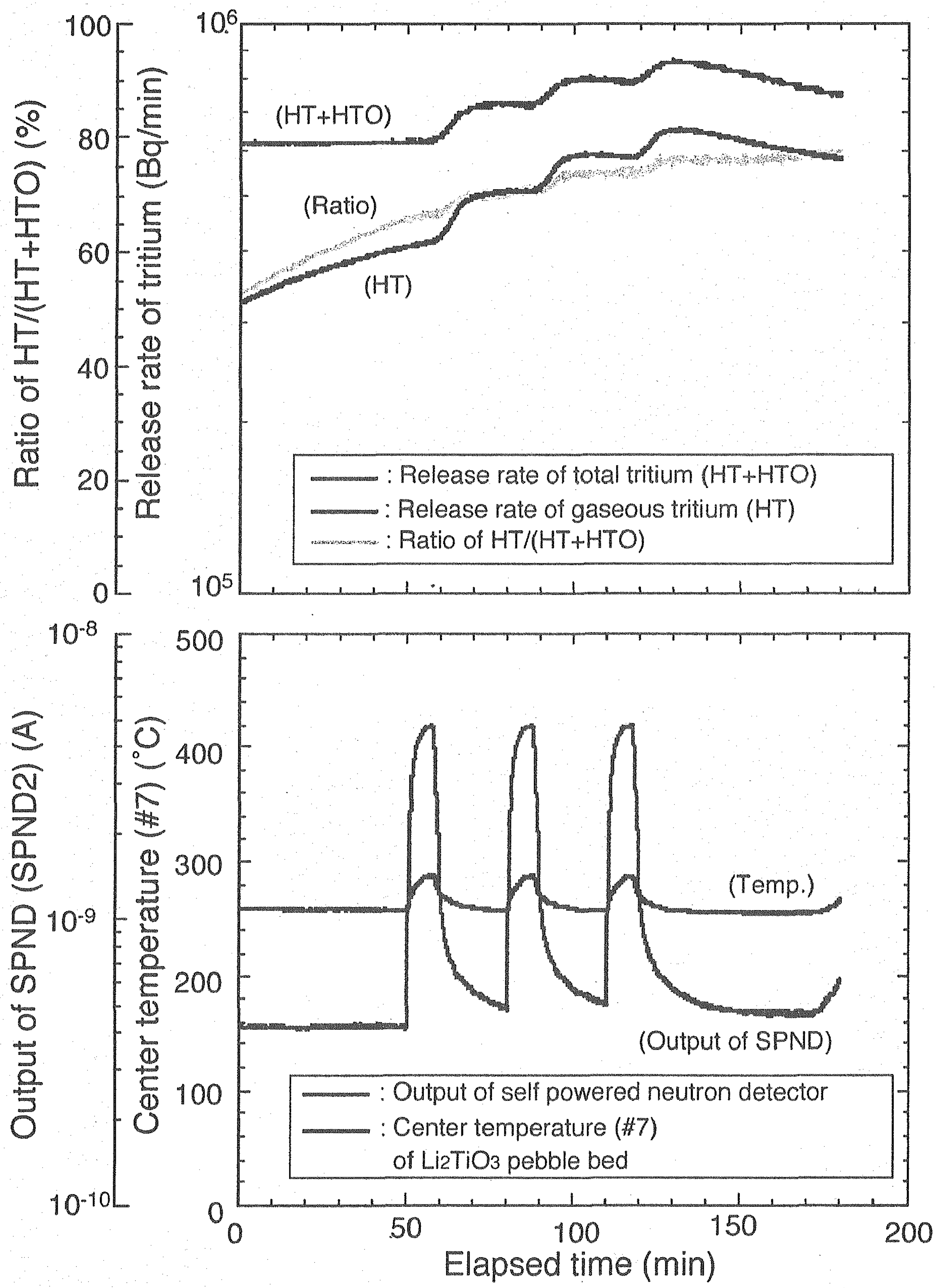


Fig.32 Result of rotating test at the reactor power up in the second irradiation test (JMTR : 30MW).

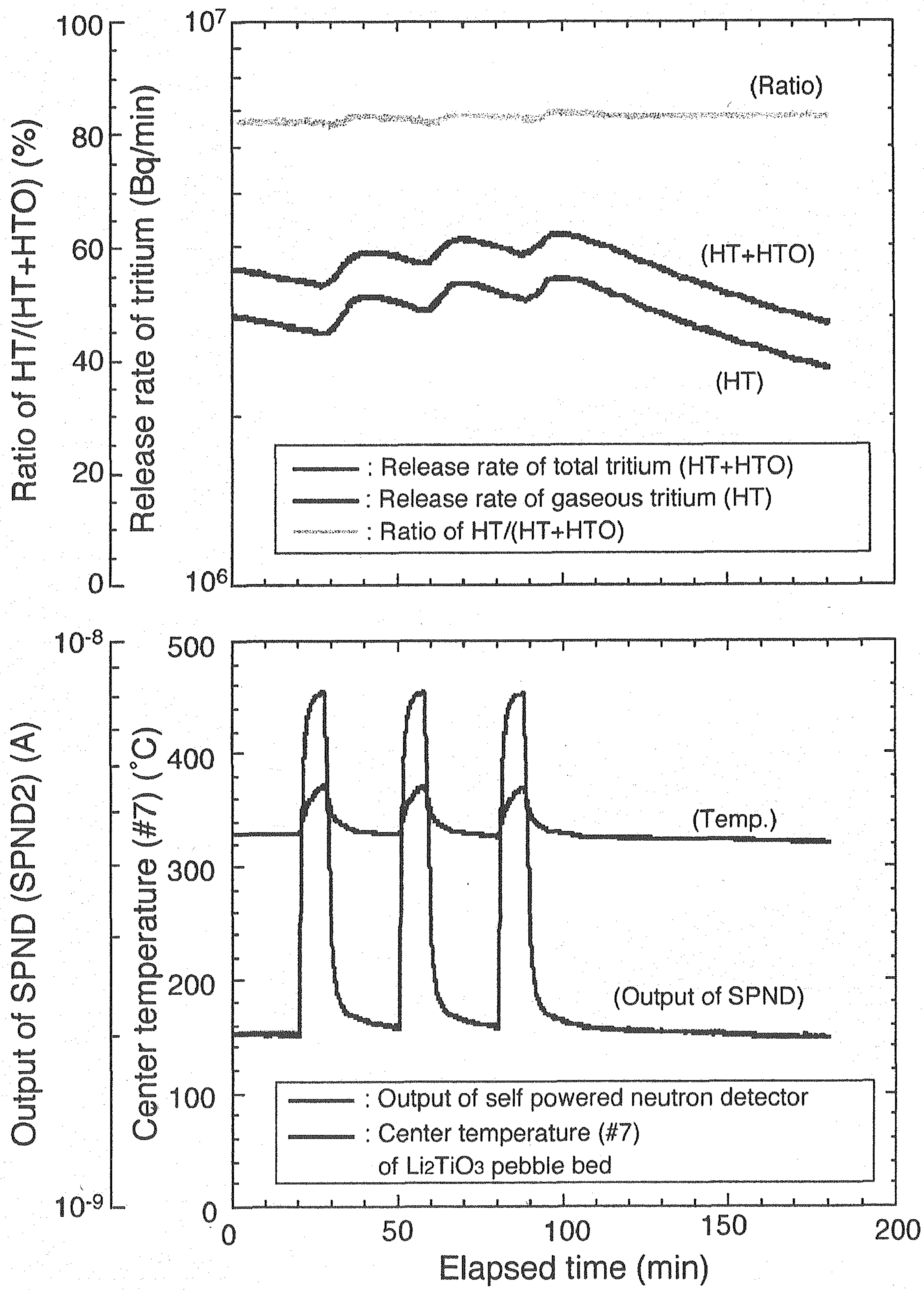
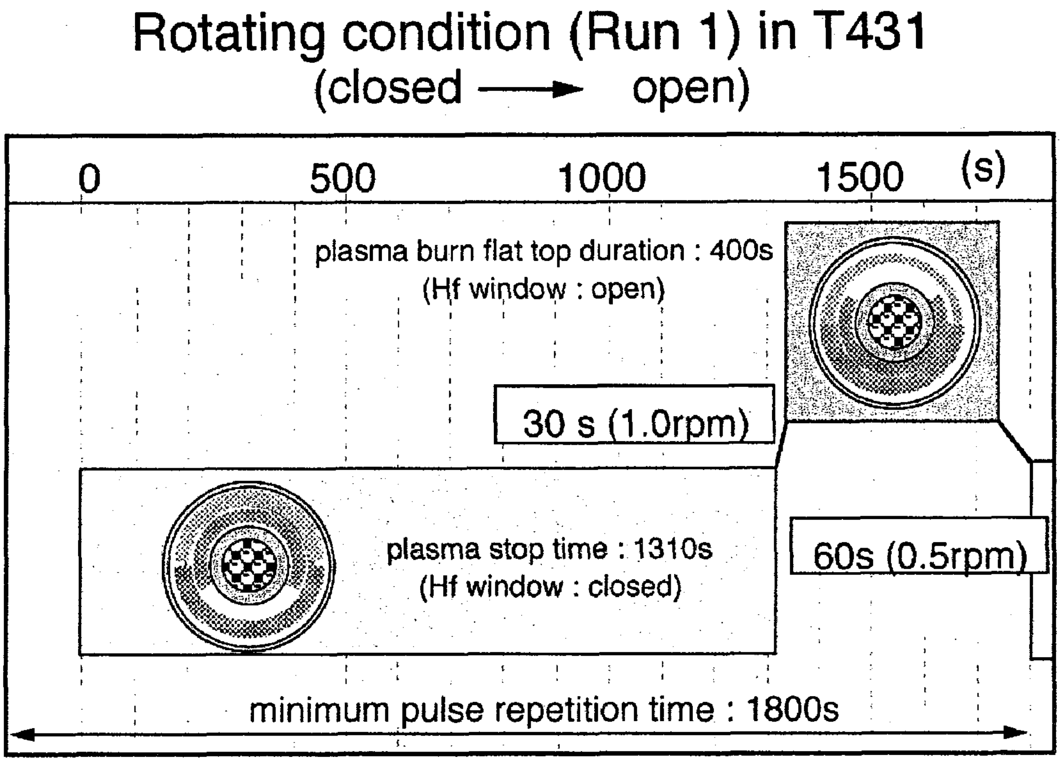


Fig.33 Result of rotating test at the reactor power up in the second irradiation test (JMTR : 45MW).



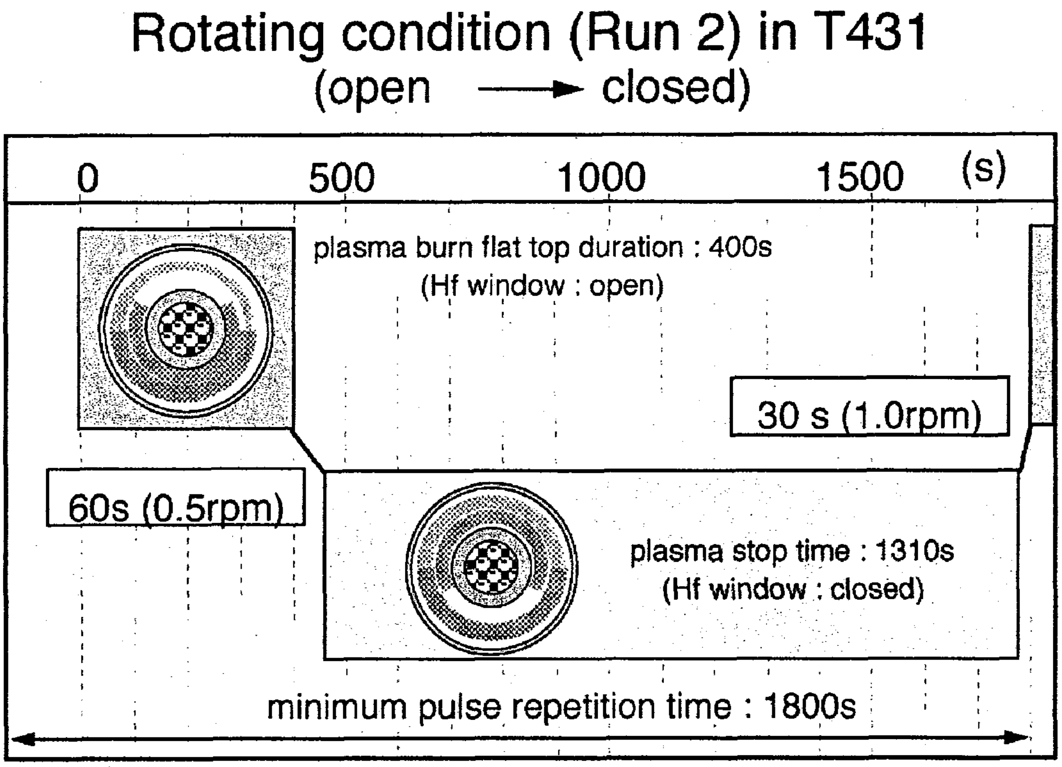


Fig.34 Conditions of ITER pulse operation test in nominal pulsed operation of ITER.

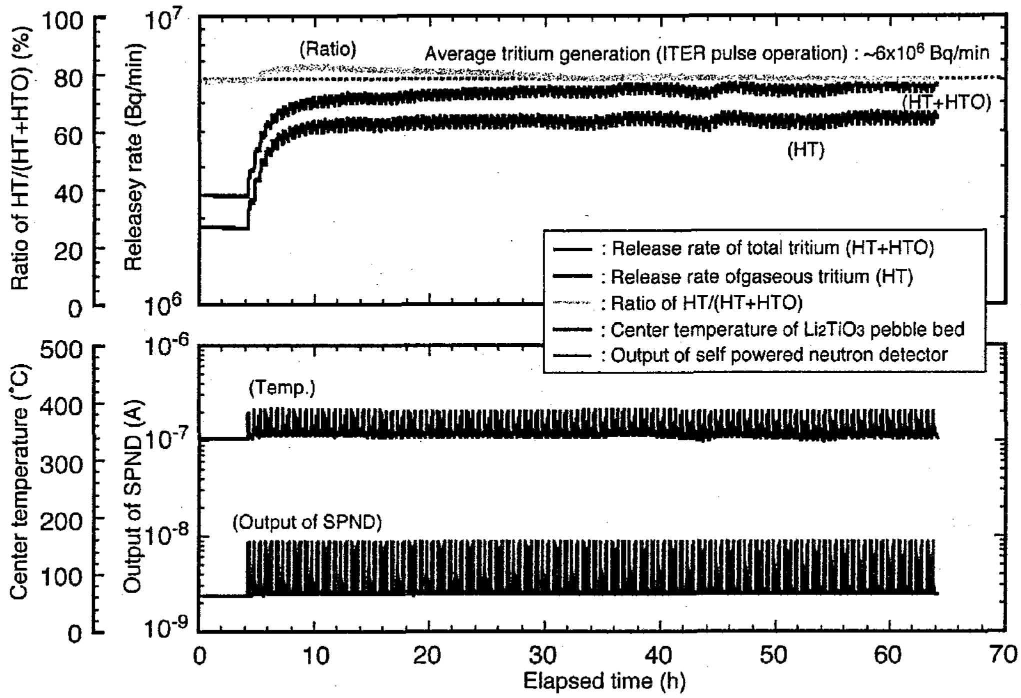


Fig.35 Result of ITER pulse operation test in the second irradiation test (Run 1).

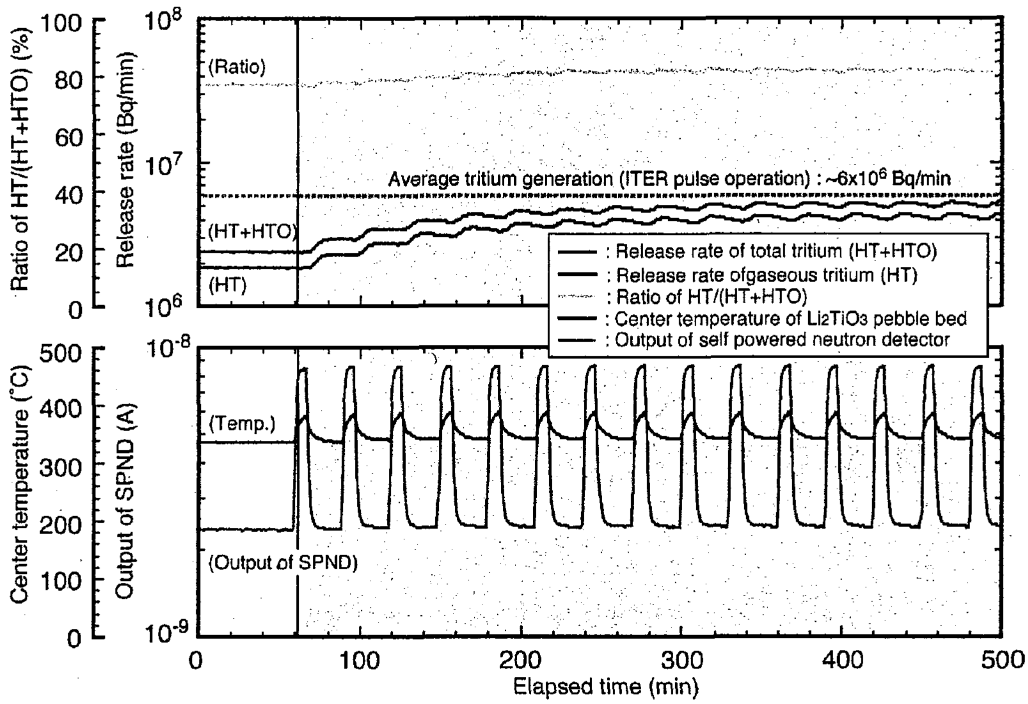


Fig.36 Result of ITER pulse operation test in the second irradiation test (Run 1-1 : star of the pulse operation).

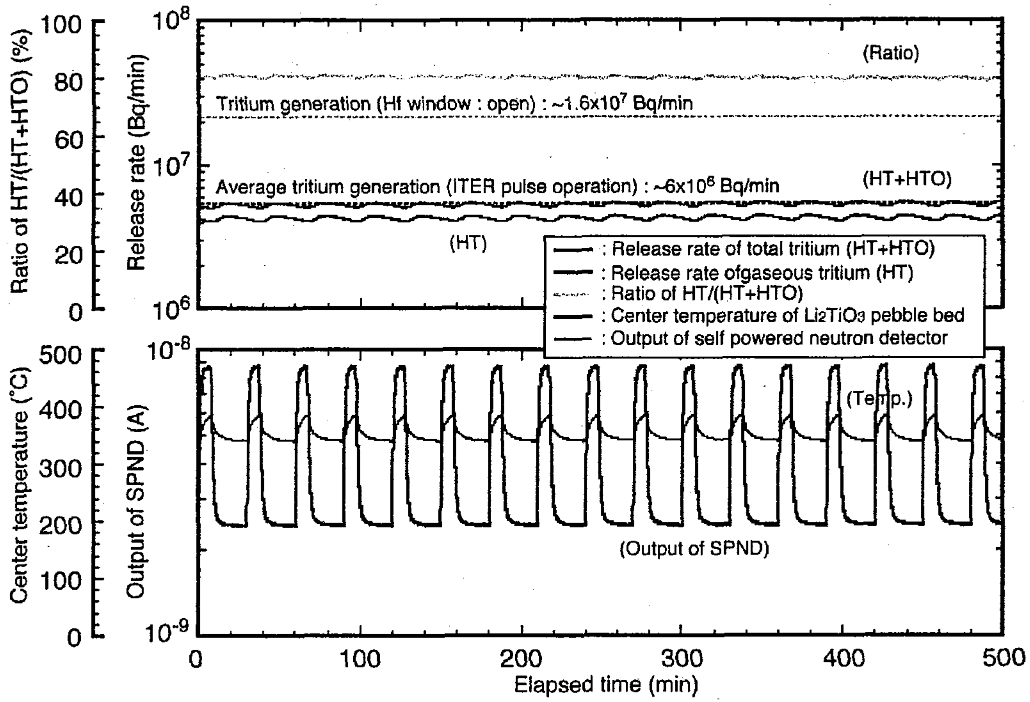


Fig.37 Result of ITER pulse operation test in the second irradiation test (Run 1-2 : after about 100 pulses).

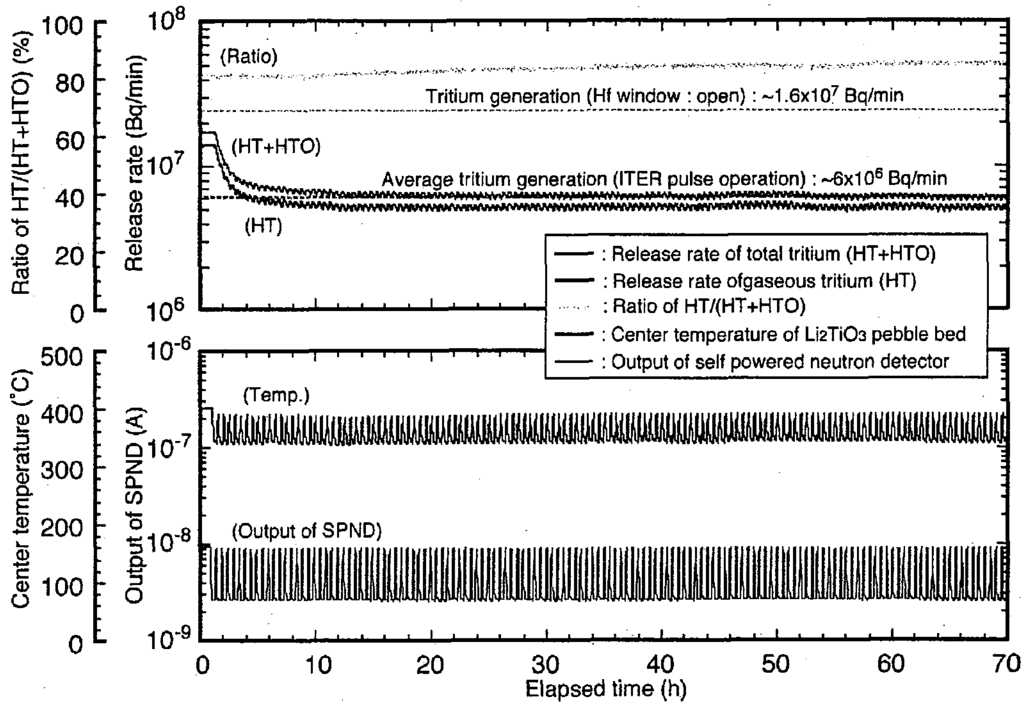


Fig.38 Result of ITER pulse operation test in the second irradiation test (Run 2).

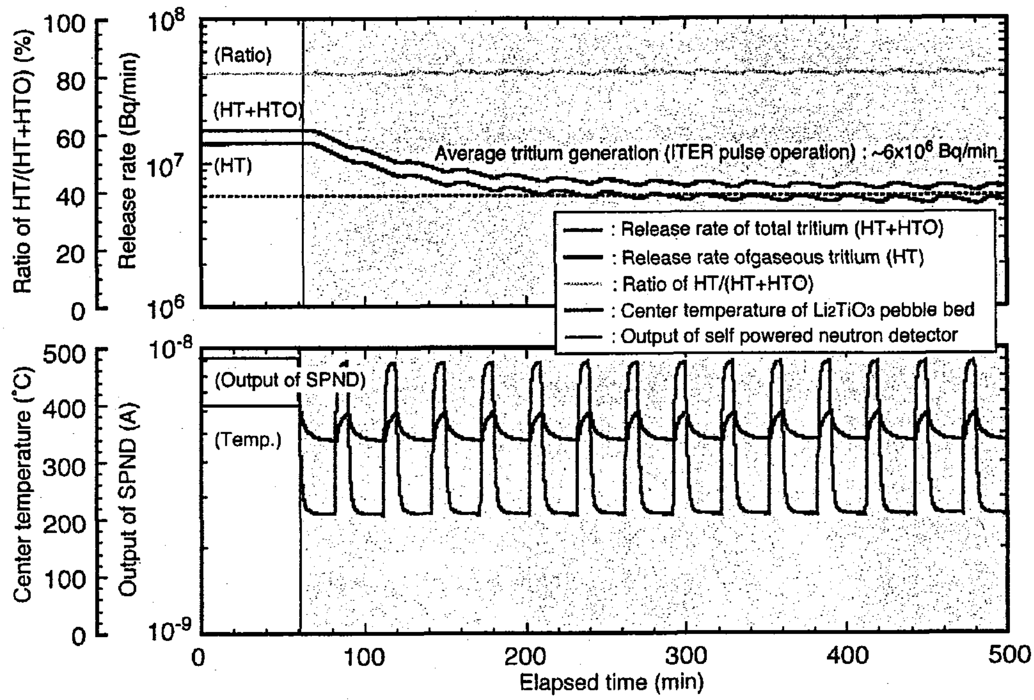


Fig.39 Result of ITER pulse operation test in the second irradiation test (Run 2-1 : start of the pulse operation).

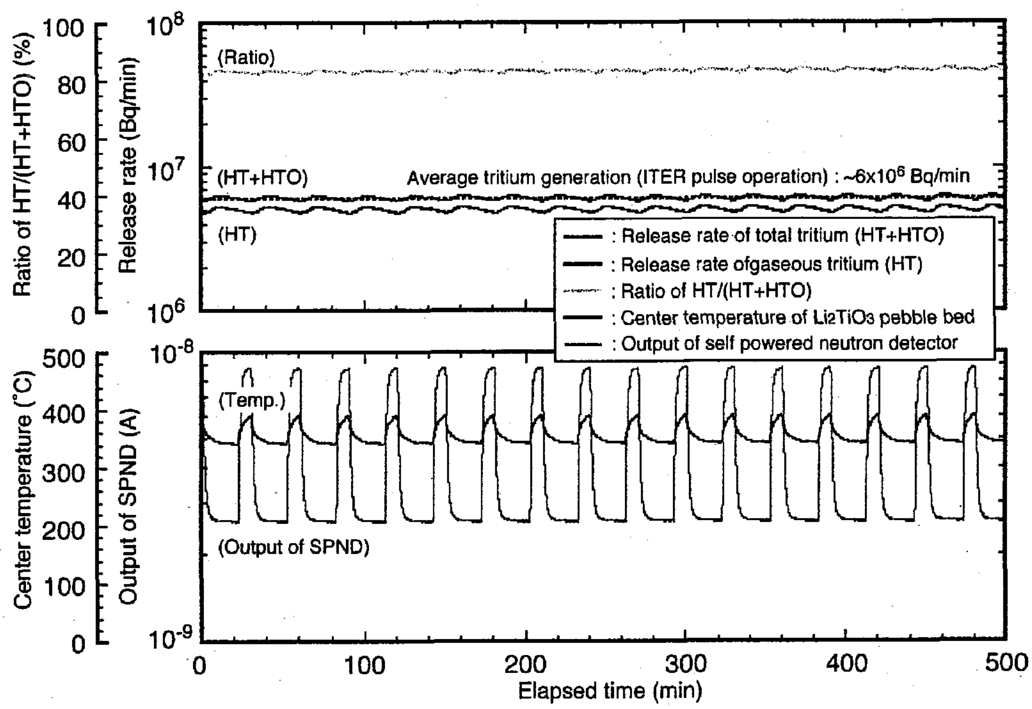


Fig.40 Result of ITER pulse operation test in the second irradiation test (Run 2-2 : after about 100 pulses).

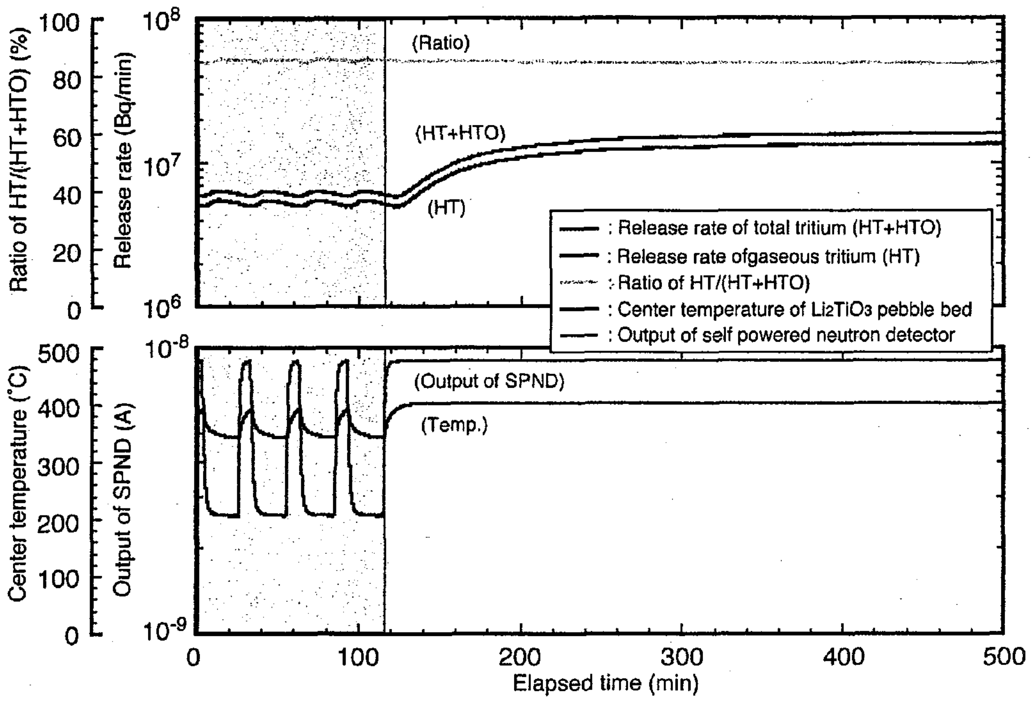


Fig.41 Result of ITER pulse operation test in the second irradiation test (Run 2-3 : after stopping the pulse operation).

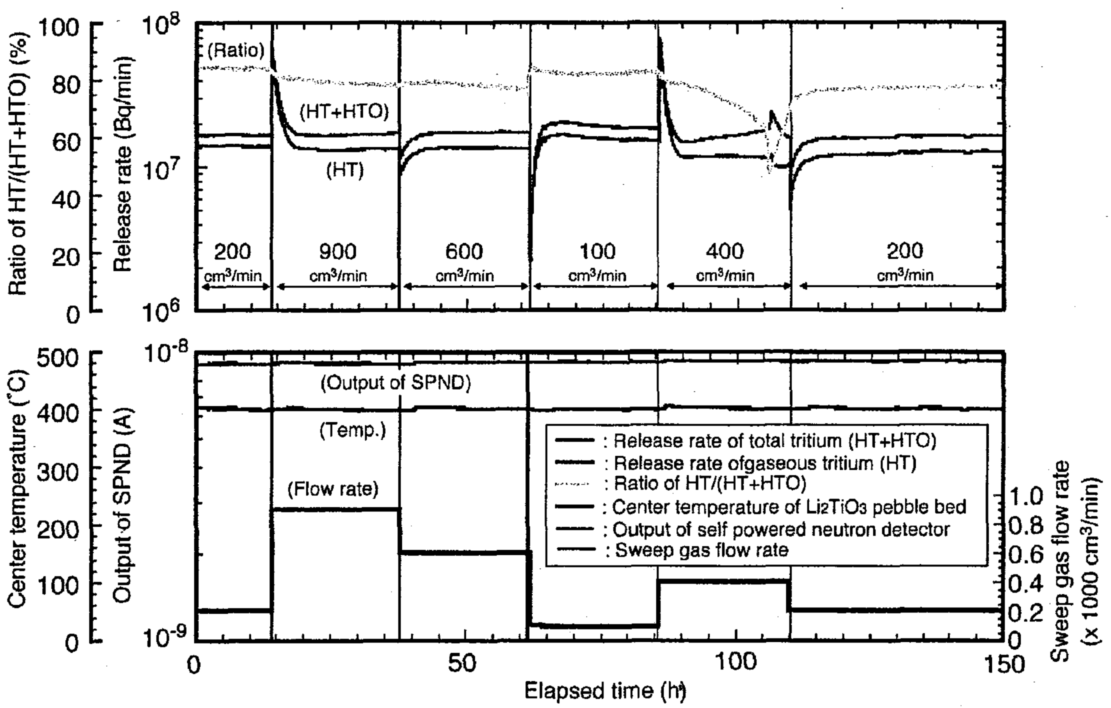
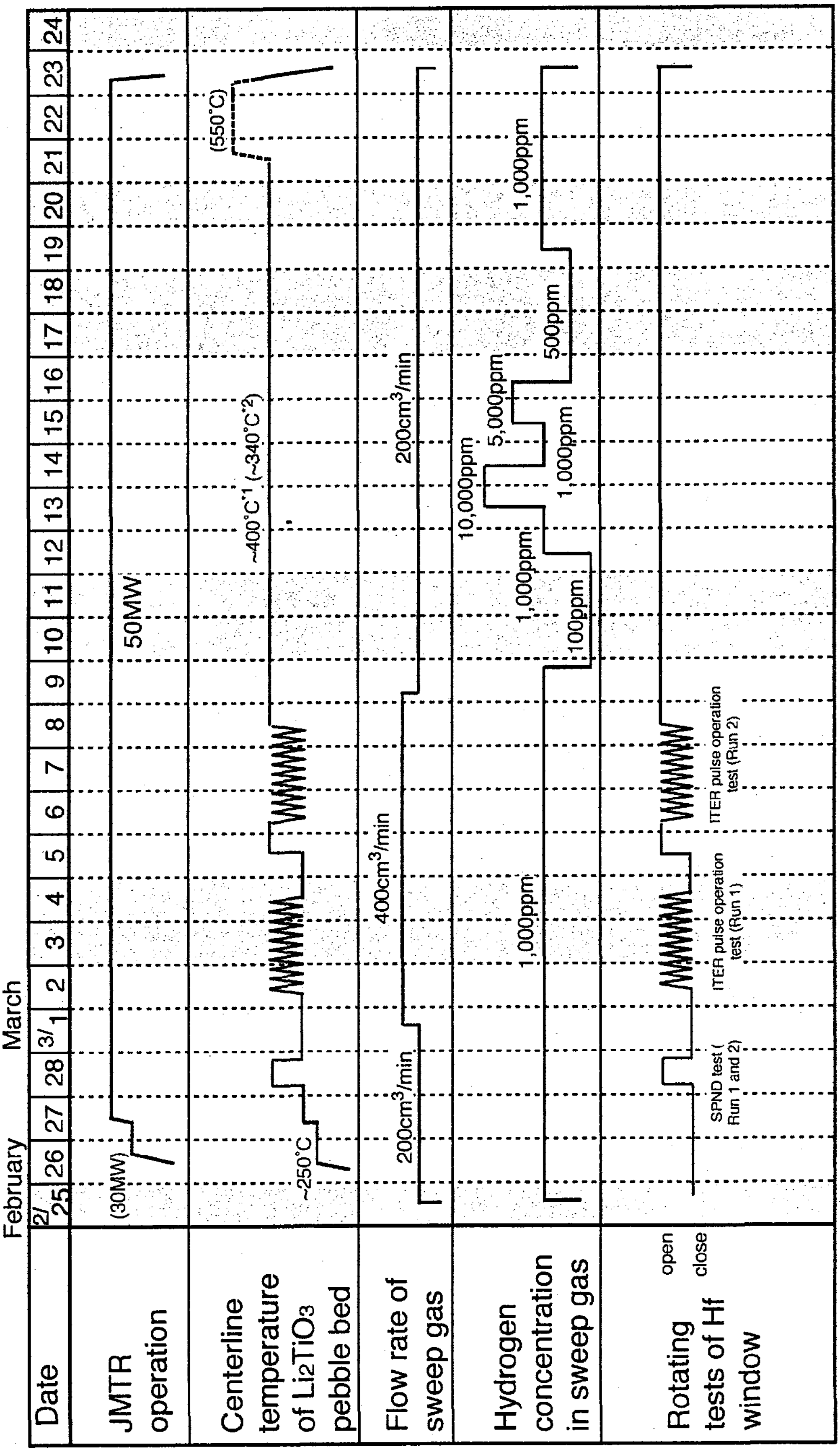


Fig.42 Result of sweep-gas flow rate changing test in the second irradiation test.



*1 : Center temperature of Li_2TiO_3 pebble bed when Hf window was opened.
*2 : Center temperature of Li_2TiO_3 pebble bed when Hf window was closed.

Fig. 43 Outline of experimental conditions on the third irradiation test.

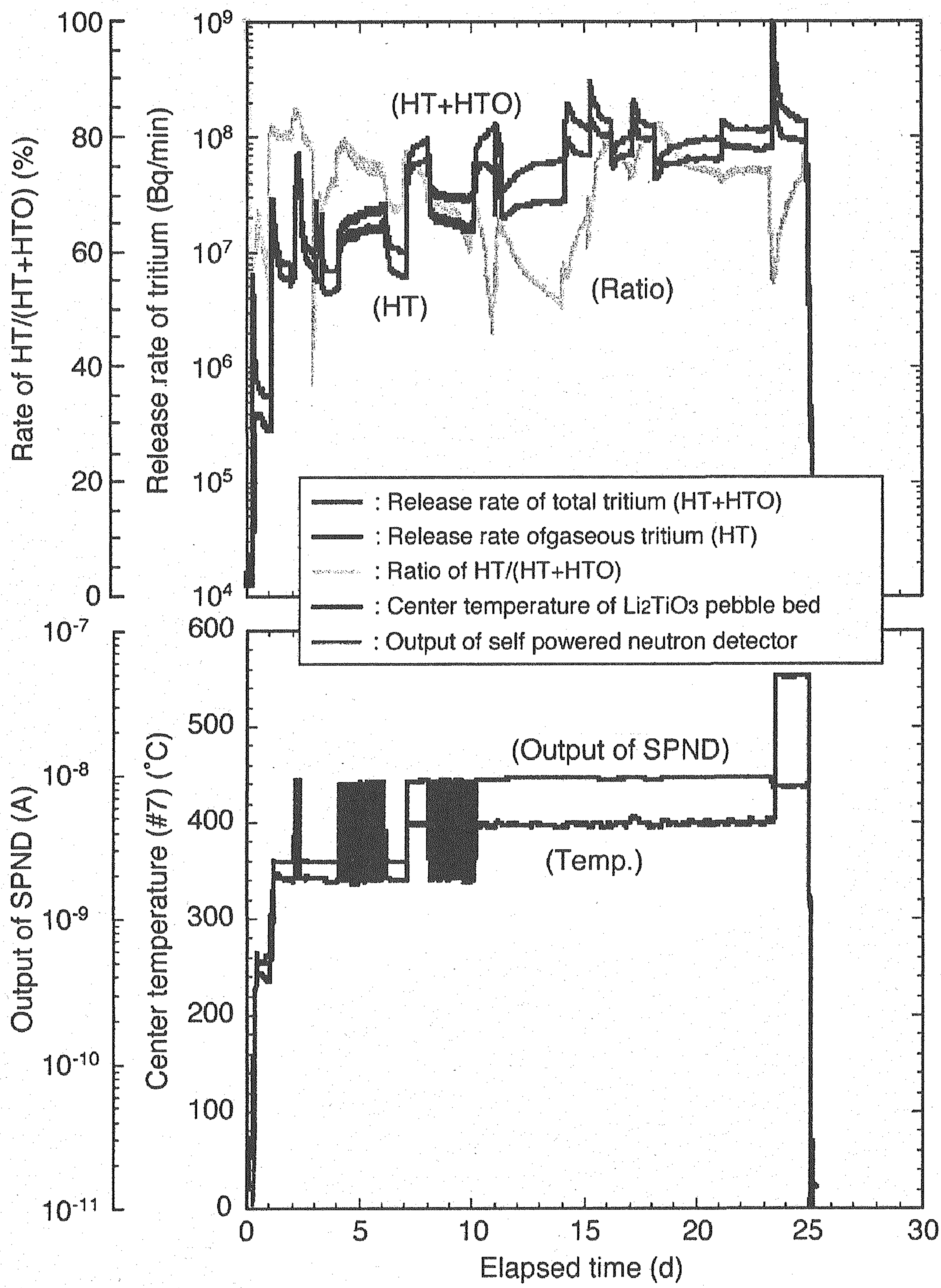


Fig.44 Result of the third irradiation test in the 138th cycle of JMTR.

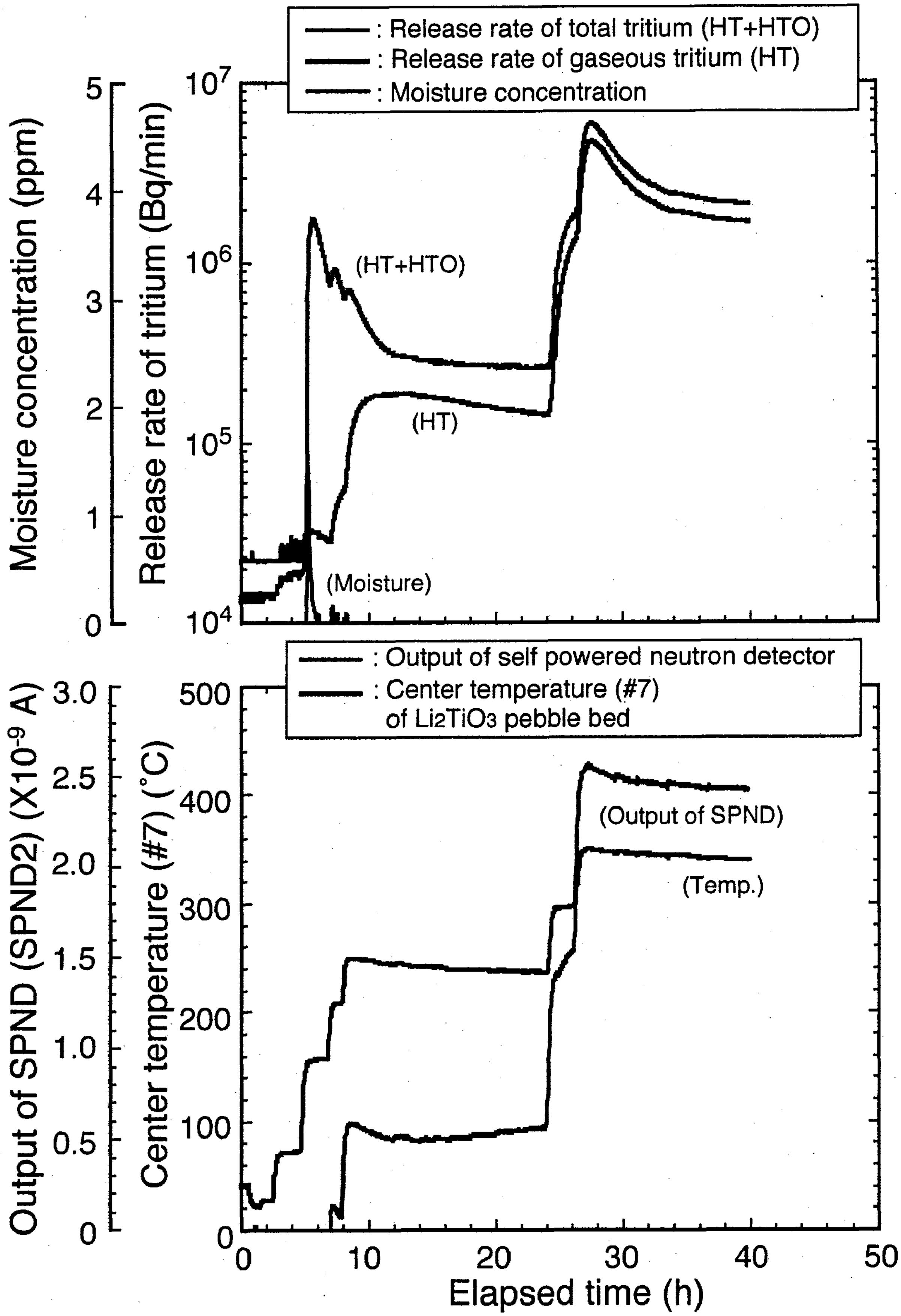


Fig. 45 Result of tritium release at the reactor startup in the third irradiation test.

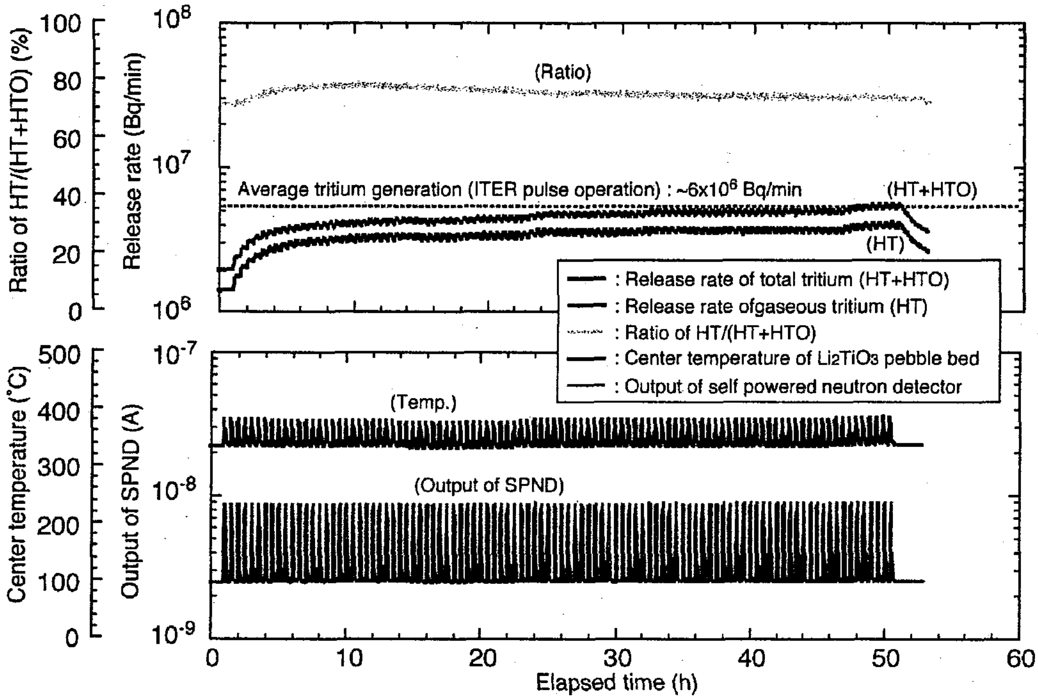


Fig. 46 Result of ITER pulse operation test in the third irradiation test (Run 1).

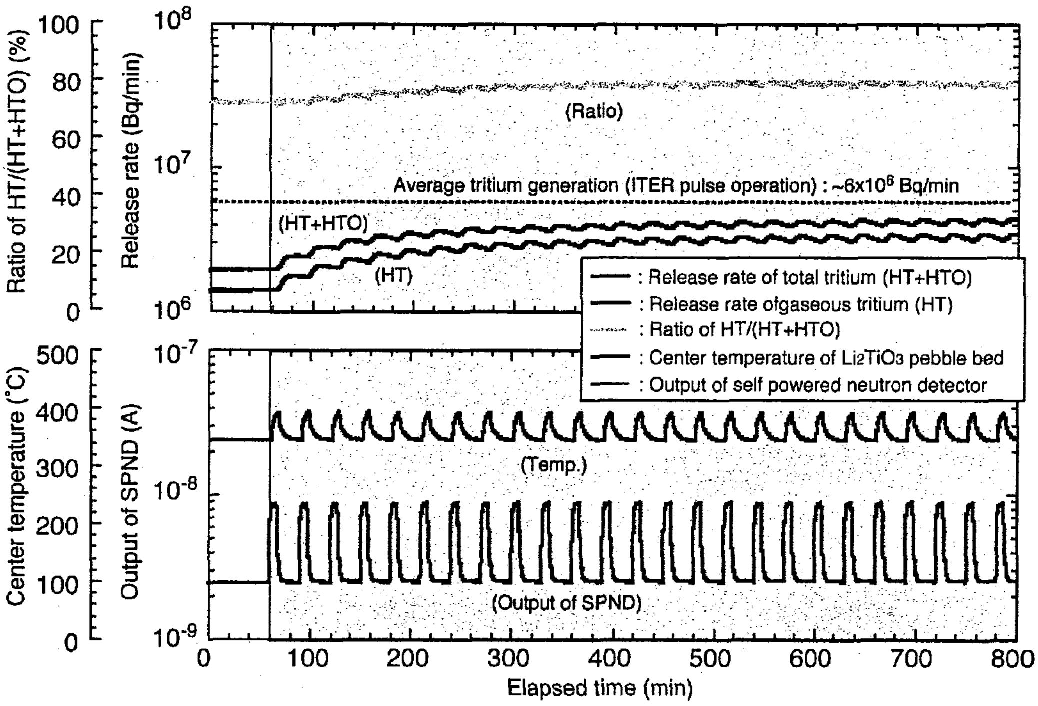


Fig.47 Result of ITER pulse operation test in the third irradiation test (Run 1-1 : star of the pulse operation).

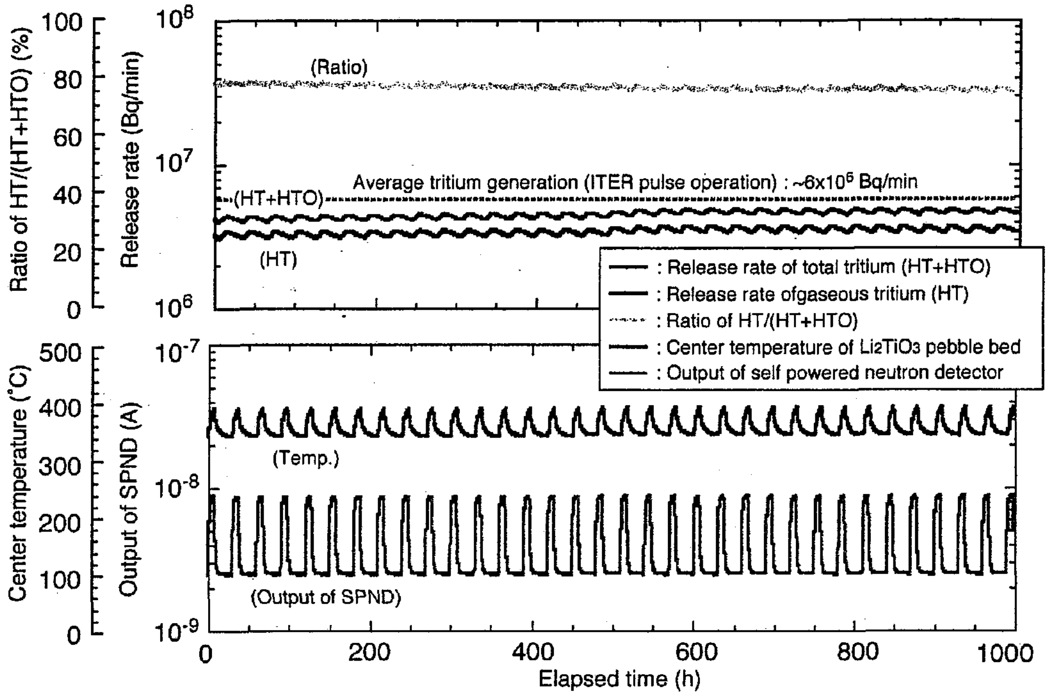


Fig.48 Result of ITER pulse operation test in the third irradiation test (Run 1-2 : after about 50 pulses).

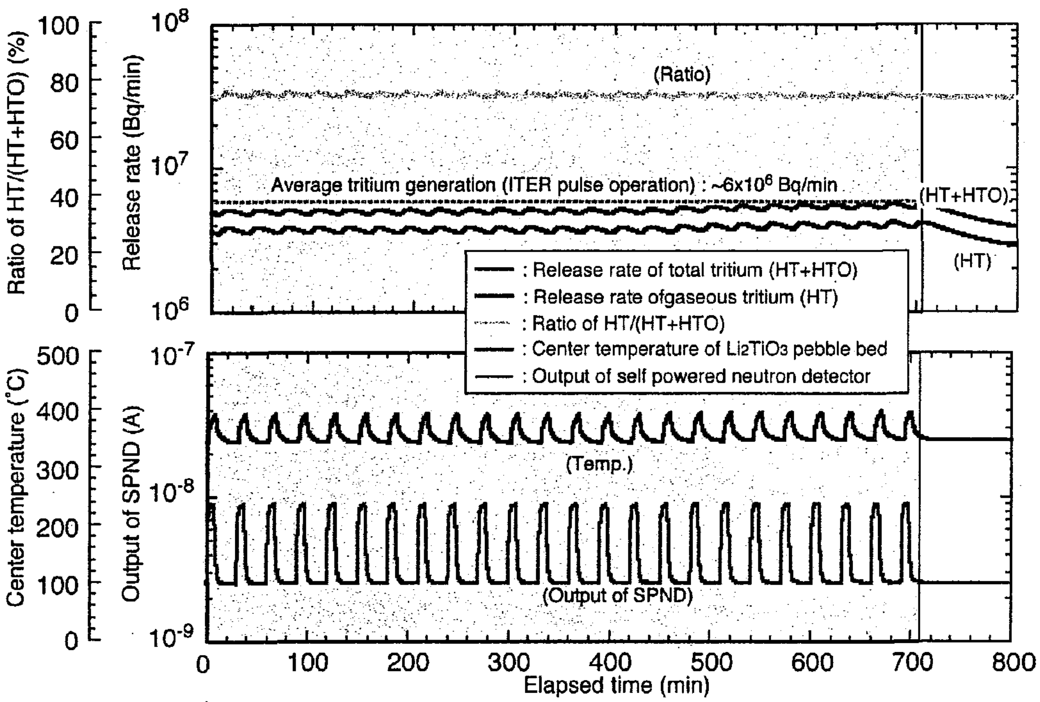


Fig.49 Result of ITER pulse operation test in the third irradiation test (Run 1-3 : after stopping the pulse operation).

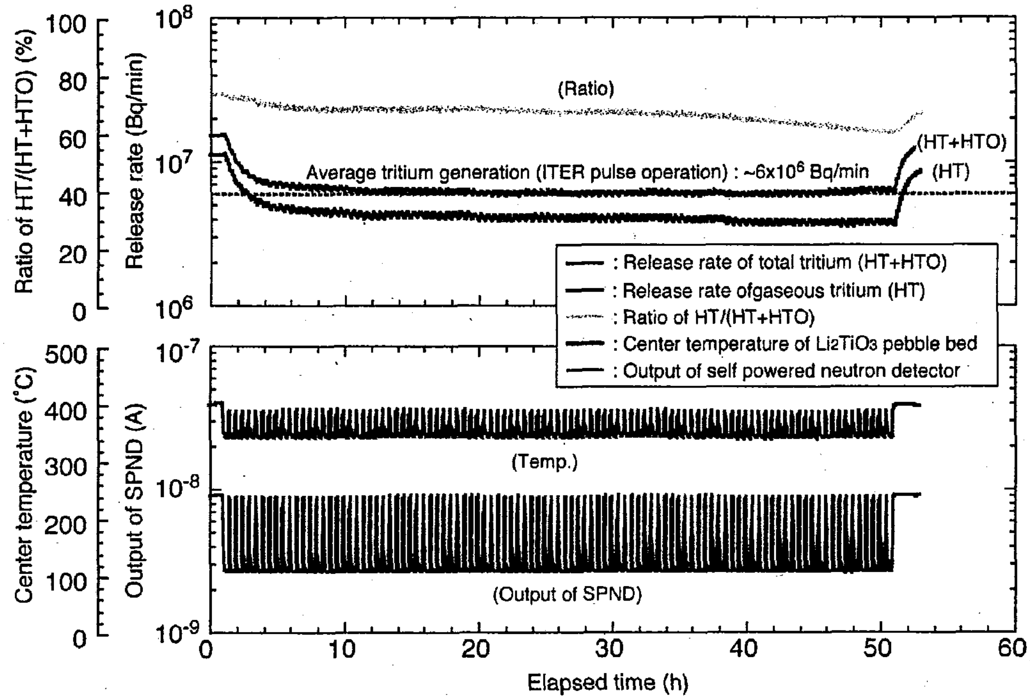


Fig.50 Result of ITER pulse operation test in the third irradiation test (Run 2).

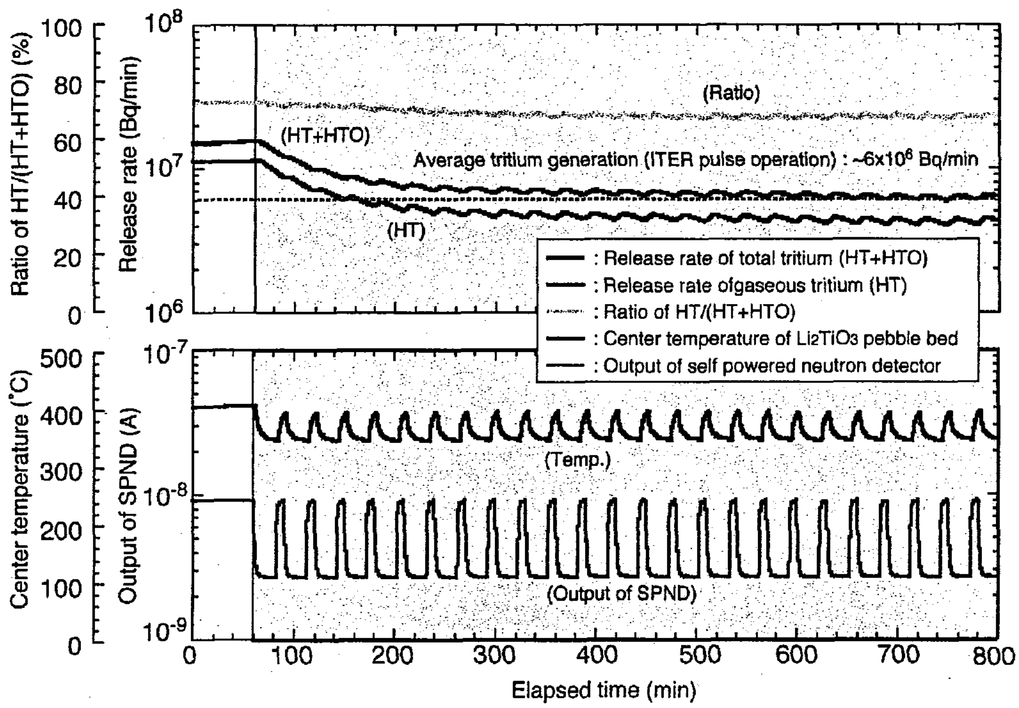


Fig.51 Result of ITER pulse operation test in the third irradiation test. (Run 2-1 : start of the pulse operation).

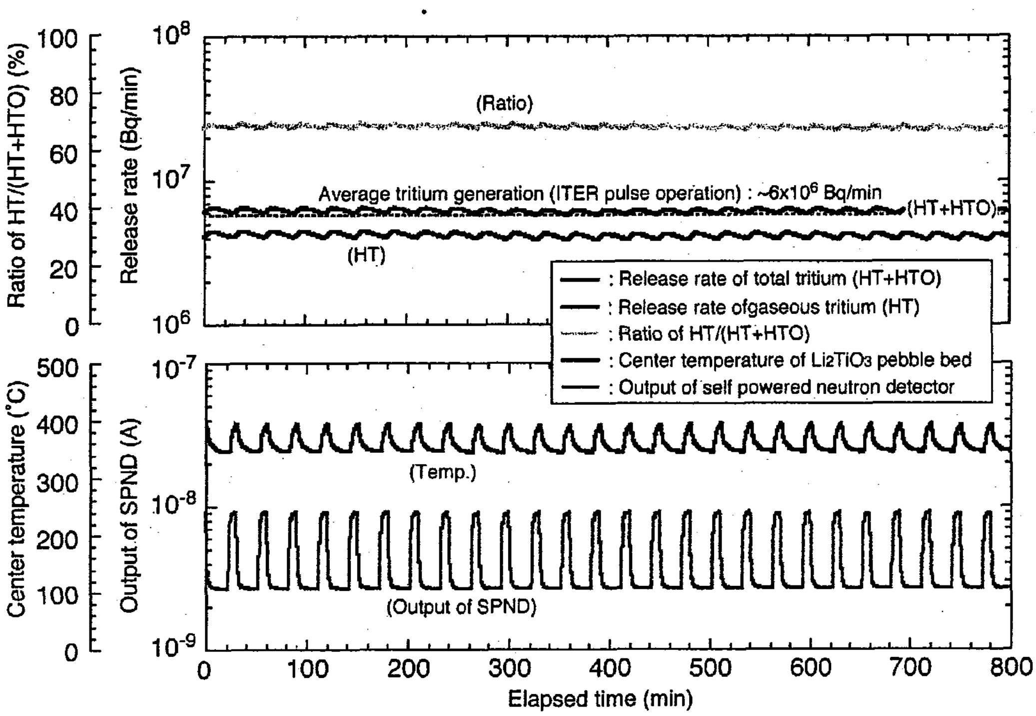


Fig.52 Result of ITER pulse operation test in the third irradiation test (Run 2-2 : after about 50 pulses).

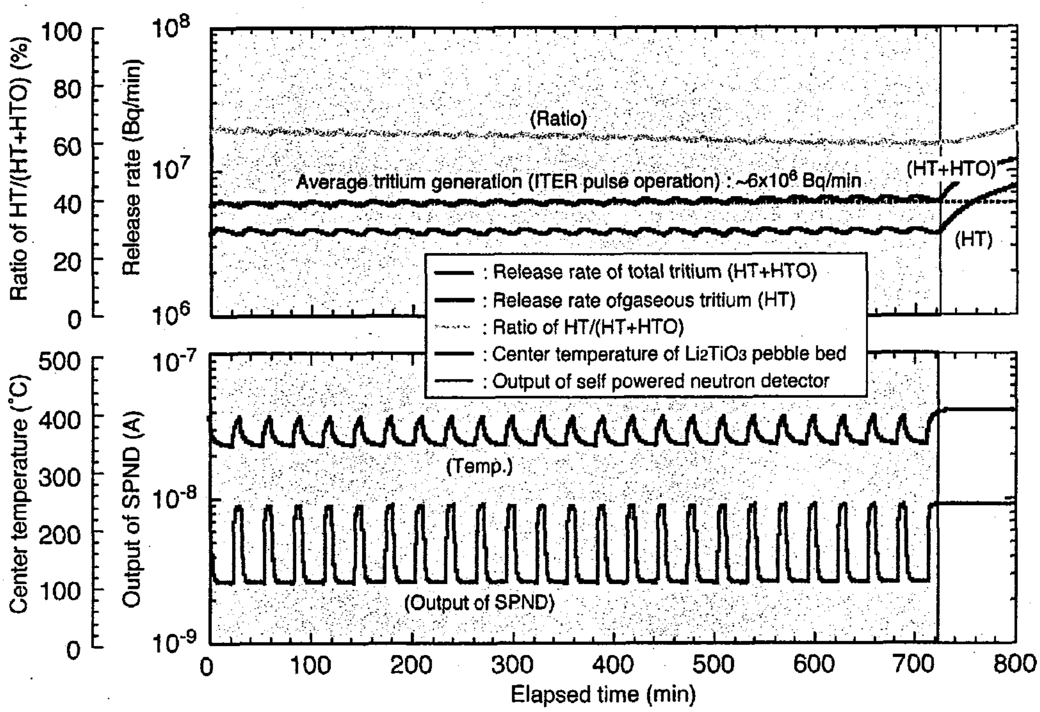


Fig.53 Result of ITER pulse operation test in the third irradiation test (Run 2-3 : after stopping the pulse operation).

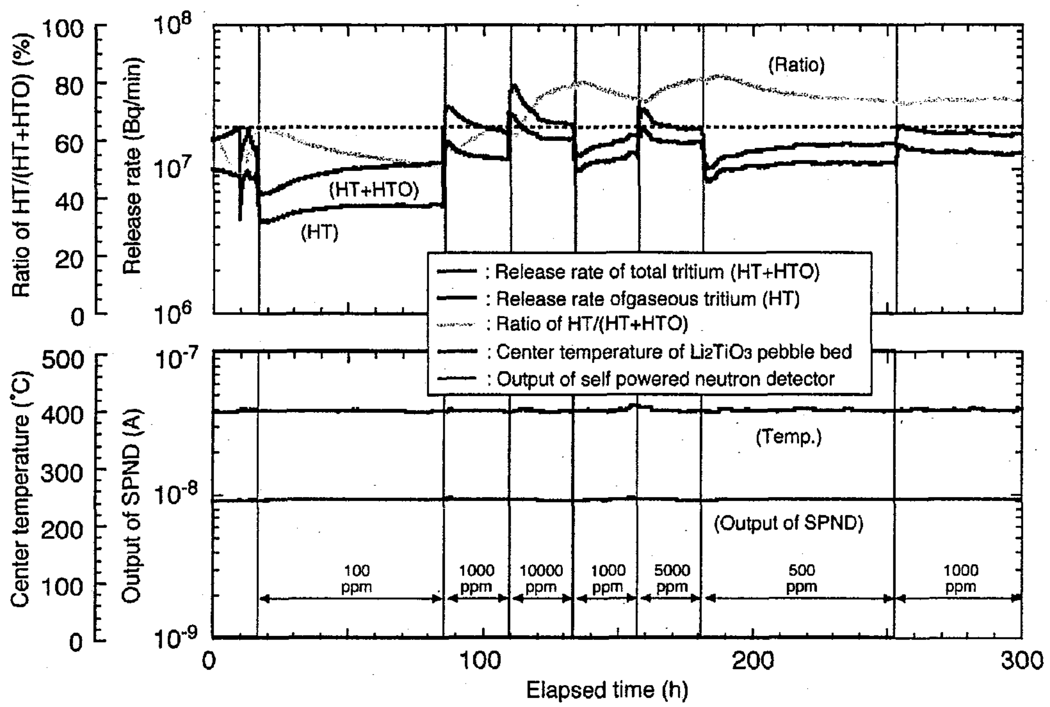


Fig.54 Result of hydrogen content changing test in the third irradiation test.

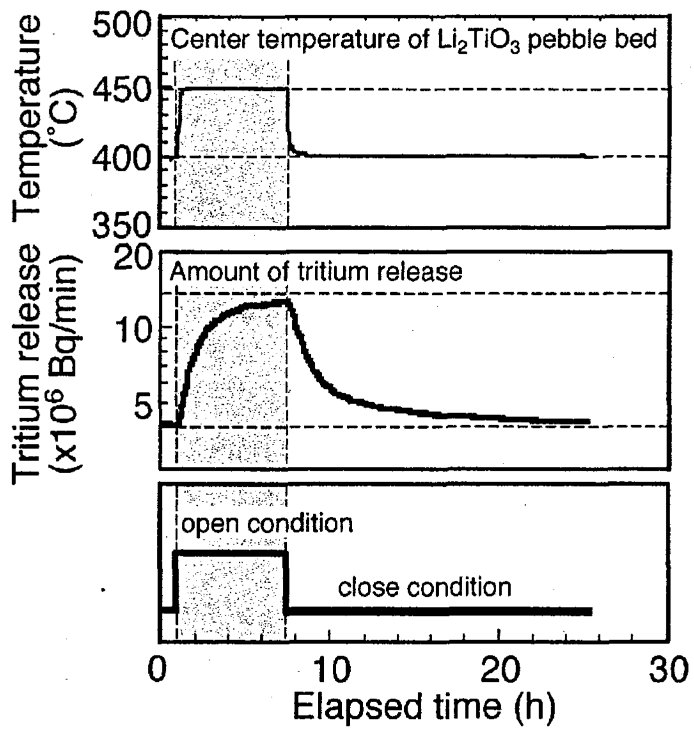


Fig.55 Result of tritium release from Li_2TiO_3 pebble bed when the window of Hf neutron absorber turned toward the reactor core (open condition) and the opposite direction of reactor core (close condition) (reprinted from "K. Tsuchiya, et al., J. Nucl. Sci. Technol. 38 (2001) 996" [13]).

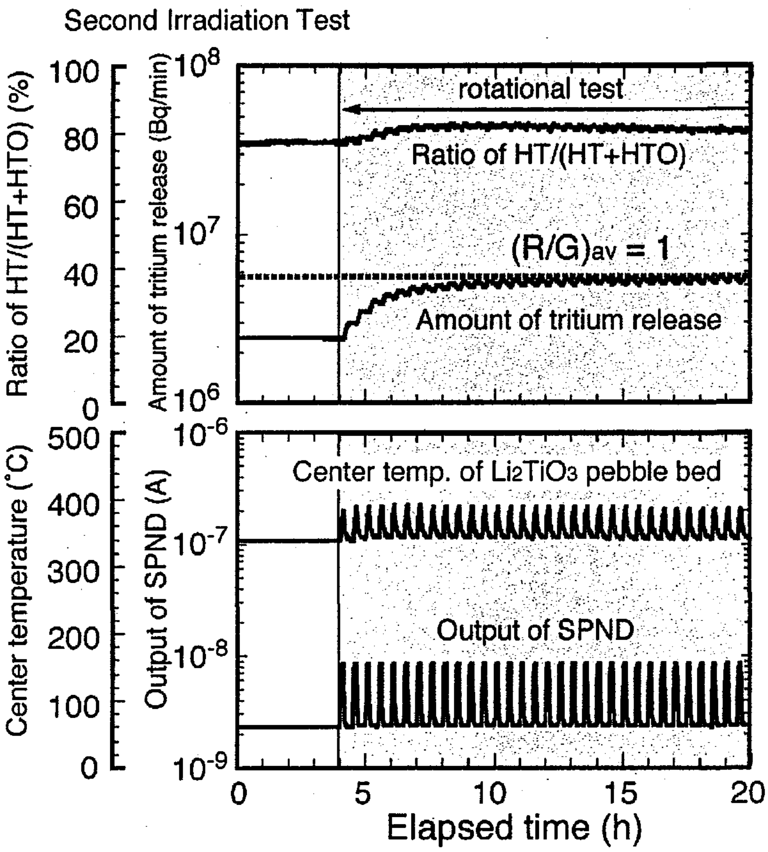


Fig.56 Result of rotation test in the condition of ITER pulsed operation (reprinted from "K. Tsuchiya, et al., J. Nucl. Sci. Technol. 38 (2001) 996" [13]).

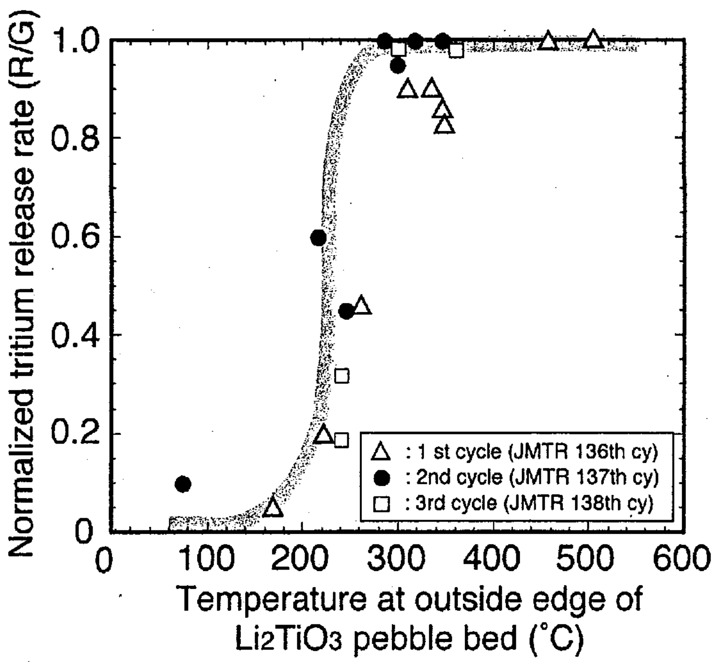


Fig.57 Relationship between the temperature at outside edge of Li_2TiO_3 pebble bed and normalized tritium release rate (R/G).

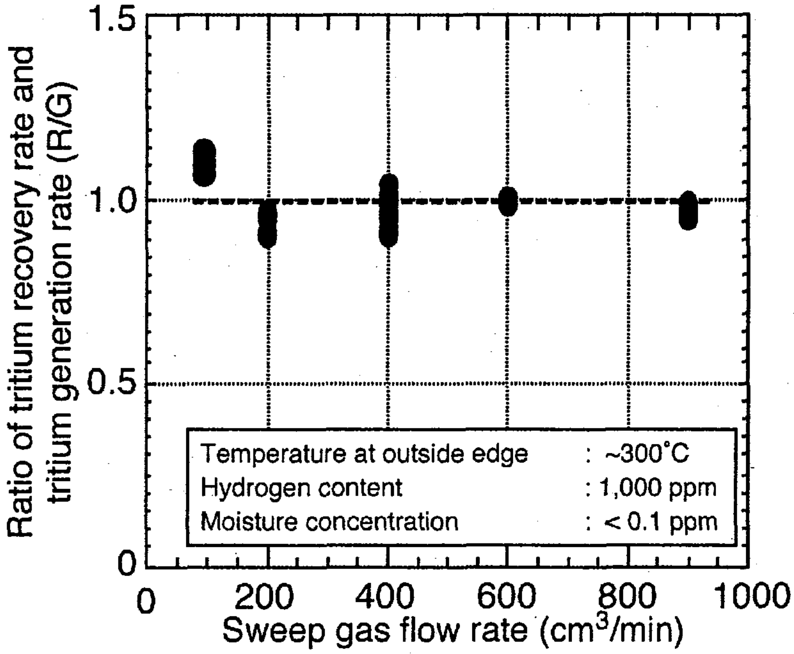


Fig.58 Result of sweep gas flow rate changing tests with the pulse-operation simulating mockup (reprinted from "K. Tsuchiya, et al., J. Nucl. Sci. Technol. 38 (2001) 996" [13]).

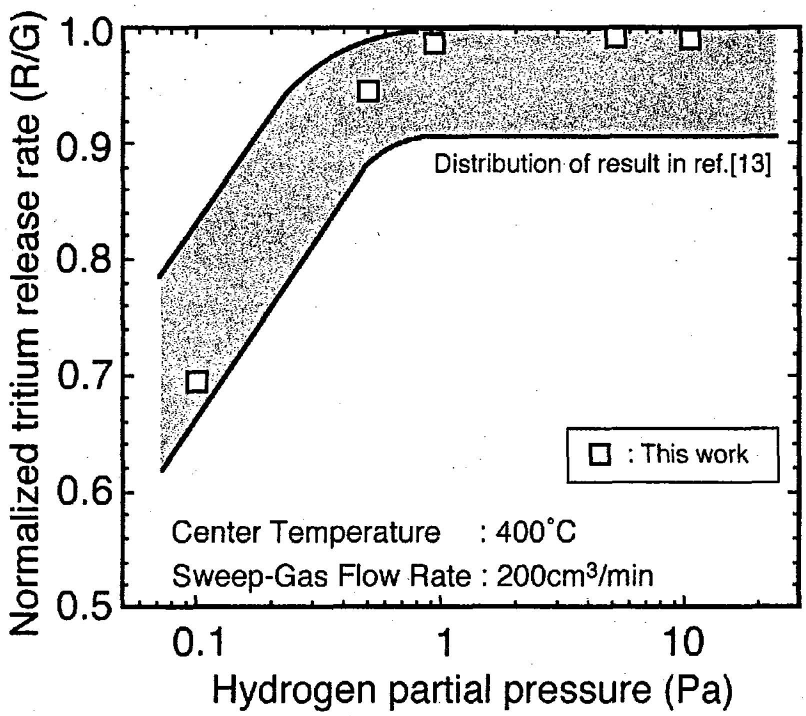


Fig.59 Relationship between hydrogen partial pressures in sweep gas and normalized tritium release rate (R/G).

Appendix A Presentation Materials for Task Meeting

Task Meeting on T431
March 14-16, Garching ITER JWS
JAERI

Breeding Blanket Development

T431 Subtask 1 - Tritium Recovery from Breeder -

H. Kawamura

The Japan Home Team

Task Objectives of T431 - Subtask 1

JAERI

The tritium recovery system from breeder blankets has been studied over the past several years, but this work is not yet completed and various issues remain.
In particular, tritium recovery at low temperature has not been clarified. Therefore in-situ irradiation data on tritium recovery should be obtained. Concerning pebble packing, the cyclic thermal behaviour of the packing fraction should be confirmed, by further study.
This task consists of two subtasks. Sub-task 1 is a phase 2 of task T365 and sub-task 2 will evaluate the pebble bed packing fraction distribution in a realistic design geometry.

Sub-Task 1: Tritium Recovery from Breeder
The objective of this task is to conduct an in-situ irradiation test in order to evaluate tritium recovery from tritium breeding material at low temperature under neutron irradiation conditions simulating ITER pulse operation.

Task Summary of T431 - Subtask 1

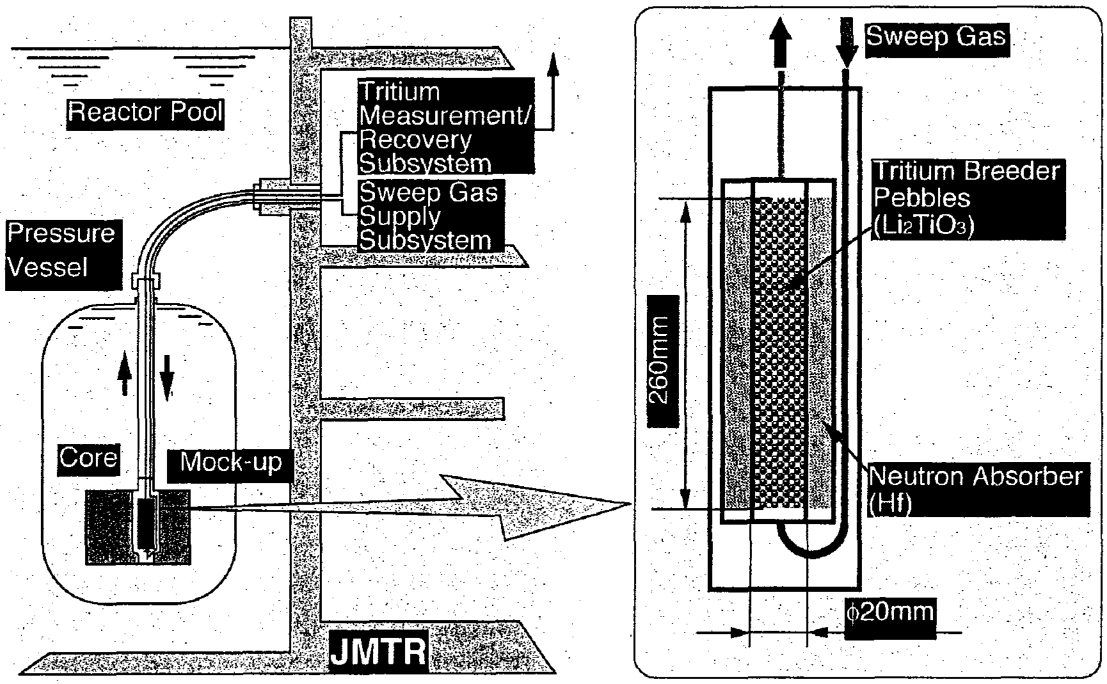
JAERI

In-situ tritium release test with lithium titanate (Li_2TiO_3) as tritium breeding material will be carried out at Japan Materials Testing Reactor (JMTR) in JAERI for a total irradiation exposure of ~75 days (3 JMTR operating cycles). The tritium breeding material is in a pebble form of ~1mm diameter and it will be irradiated in the temperature range of 200 to 400 °C. The tritium released from Li_2TiO_3 will be continuously monitored, as the irradiation temperature, purge gas parameters and neutron flux are changed.

Irradiation Test of Tritium Breeder with JMTR

- Outline of Irradiation System -

JAERI



Task Description of T431 - Subtask 1 (1)

JAERI

In-situ data on tritium recovery from tritium breeding material at low temperature are indispensable for the design of the ITER breeding blanket. This test will be carried out under transient neutron irradiation to simulate ITER pulse operation with JMTR, in order to obtain in-situ data on tritium recovery at low temperature.

Irradiation Capsule

- Packing Weight of Tritium Breeding Material : ~130 g
- Packing Fraction of Tritium Breeding Material : ~80%
- Structure material : SS316
- Neutron Absorber : Hafnium (Hf)

Tritium Breeding Material

- Material : Li_2TiO_3
- Shape : Pebble
- Density : ~80 %T.D.
- 6Li Enrichment : natural
- Grain Size : ~5μm

Task Description of T431 - Subtask 1 (2)

JAERI

Irradiation Conditions

- Reactor : JMTR
- Irradiation Hole : M-2
- Irradiation Period : 3 cycles in JMTR (1 cycle = ~25 days)
- Neutron Flux
  - Thermal Neutron (E<0.683eV) : 3×10^{11} ~2×10^{12} n/cm²/s (TBD)
  - Fast Neutron (E>1.0 MeV) : ~5×10^{11} n/cm²/s (TBD)
- Tritium Generation Rate : ~ 5 Ci/d (TBD)
- Irradiation Temperature (Tritium breeder region) : 200~400 ° C
- Volume Heating Rate : ~2 W/cm³ (TBD)
- Changing width of neutron flux : one order

Task Description of T431 - Subtask 1 (3)

JAERI

Sweep Gas Conditions

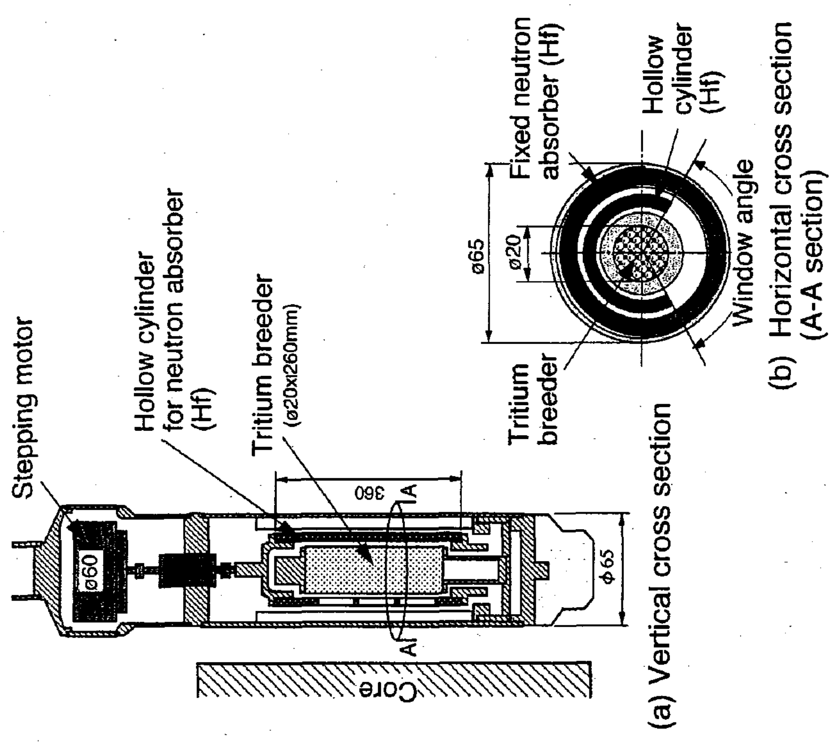
- Sweep Gas : pure He
  : $He+H_2$
- Gas Flow Rate : 10 ~ 1000 cm^3/min
- Hydrogen Content in Sweep Gas : 0 ~ 10000 ppm

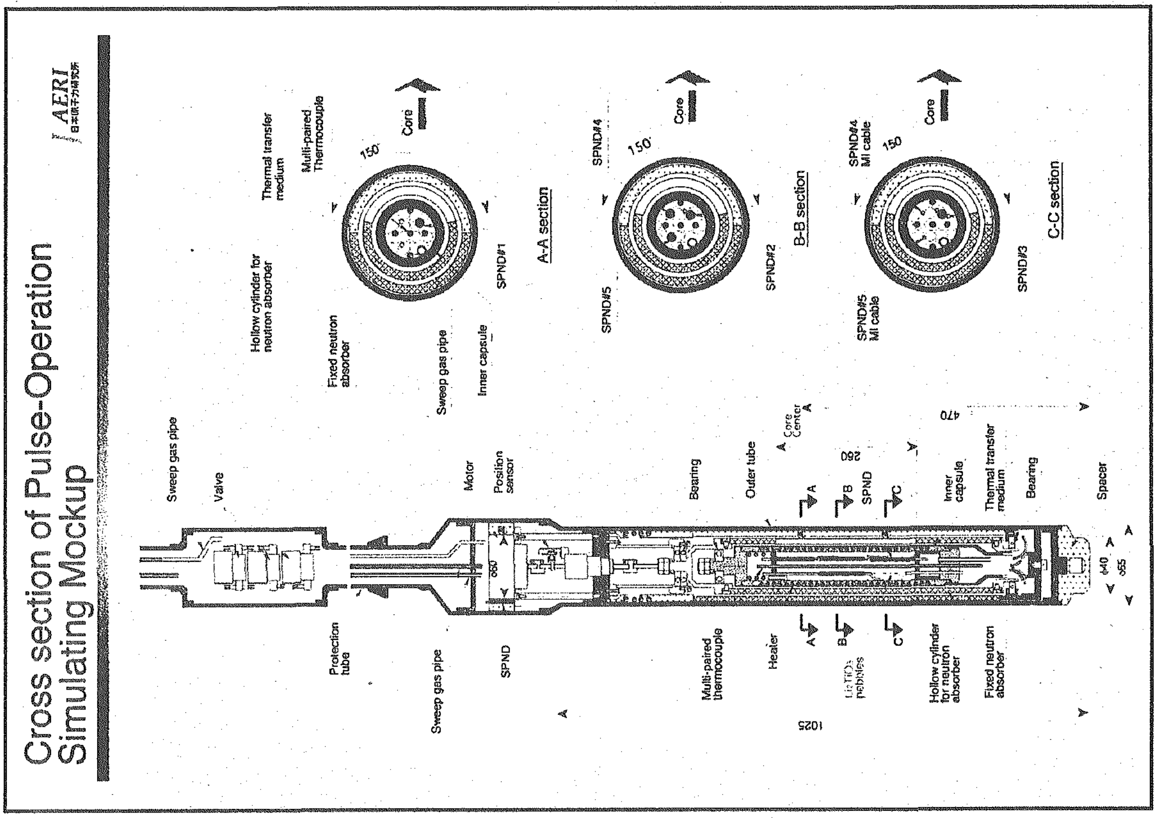
Tritium Release Test

- Change of Sweep Gas Flow Rate
- Change of Hydrogen Content in Sweep Gas
- Change of Irradiation Temperature and Neutron Flux

Conceptual Design on Pulse-Operation Simulating Mockup

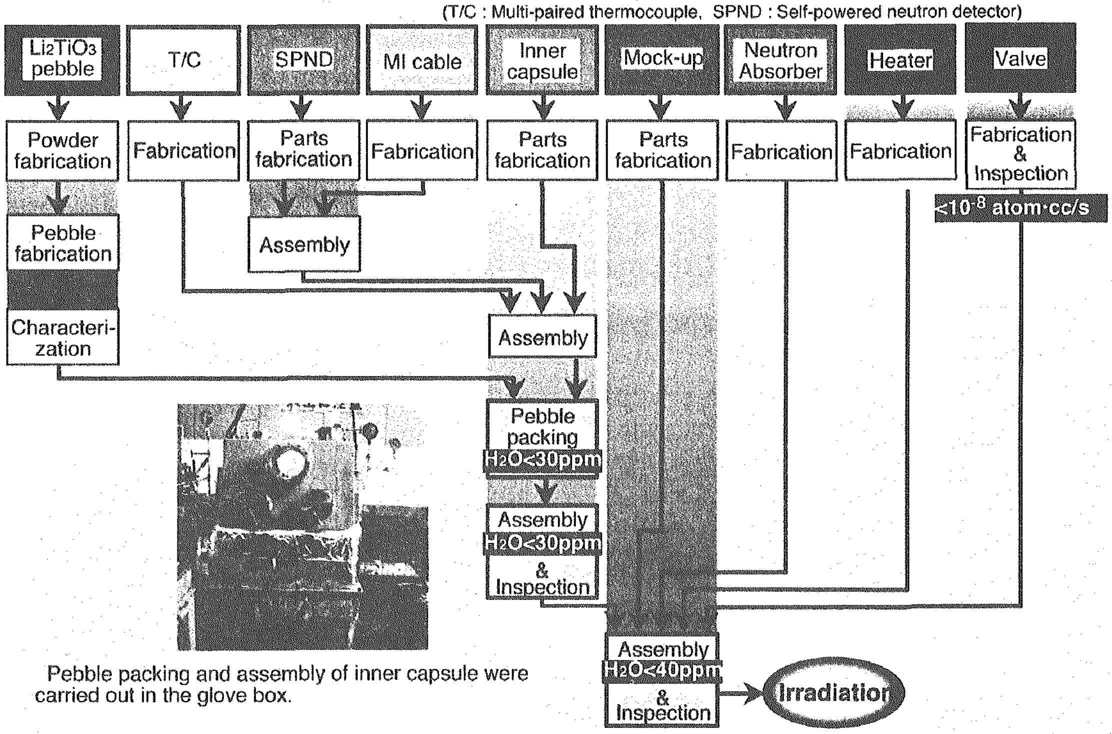
JAERI





Outline of Fabrication Flow of Mock-up

(T/C : Multi-paired thermocouple, SPND : Self-powered neutron detector)



Pebble packing and assembly of inner capsule were carried out in the glove box.

Outline of Pebble Fabrication by Wet Process

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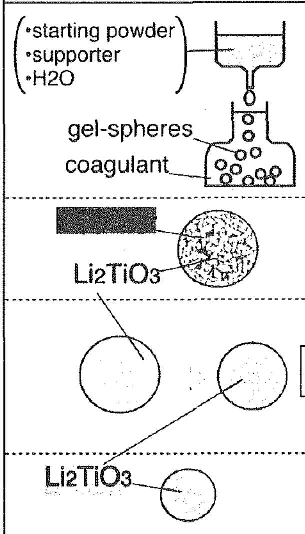

Fabrication Process	Wet process with dehydration reaction	Wet process with substitution reaction
Mixing	supporter : PVA*1 coagulant : acetone	supporter : ammonium alginate*1 coagulant : zinc chloride*1
Dropping	coagulant Temp. : <-20°C	coagulant Temp. : RT
Drying	drying time : ~24 h	drying time : ~0.5 h
Calcination	in air ~650°CX6h	reduction (H_2 gas) ~1000°CX4h
Sintering	D_{min}*2=0.5mm	D_{min}*2=~0.2mm

*1 : candidate material, *2 : minimum diameter of pebbles with high sphericity

Specification of Li_2TiO_3 Pebbles

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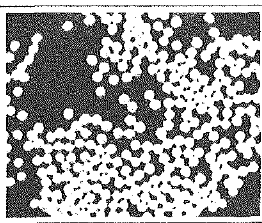
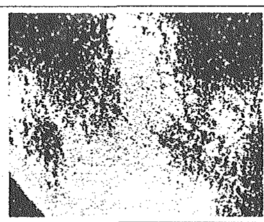
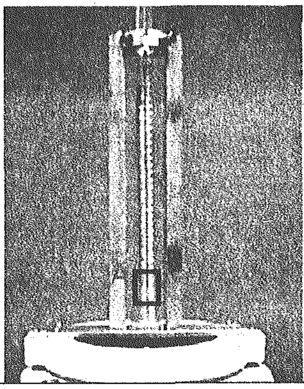
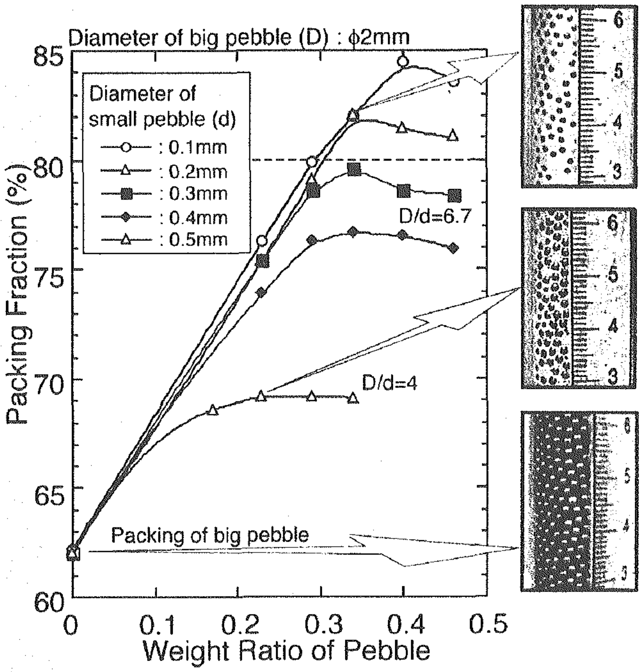
properties \ pebbles	**Large Pebbles**	**Small Pebbles**
Material	Li_2TiO_3	Li_2TiO_3
Fabrication Method	Wet process with dehydration reaction	Wet process with substitution reaction
Diameter	ϕ1.7~ϕ2.36 mm (~ϕ1.9 mm av.)	ϕ0.25~ϕ0.3 mm (~ϕ0.27 mm av.)
Density	2.86 g/cm^3	2.82 g/cm^3
Sphericity	1.07	1.11
Grain Size	<5 µm	<5 µm
Collapse load	73.4 N	4.57 N
Impurity (ppm)	Ca:<2, Na:82, Al:9, Si:14	Ca:18, Na:39, Al:23, Si:73
Photograph		

Relationship between Diameter of Pebble and Packing Fraction

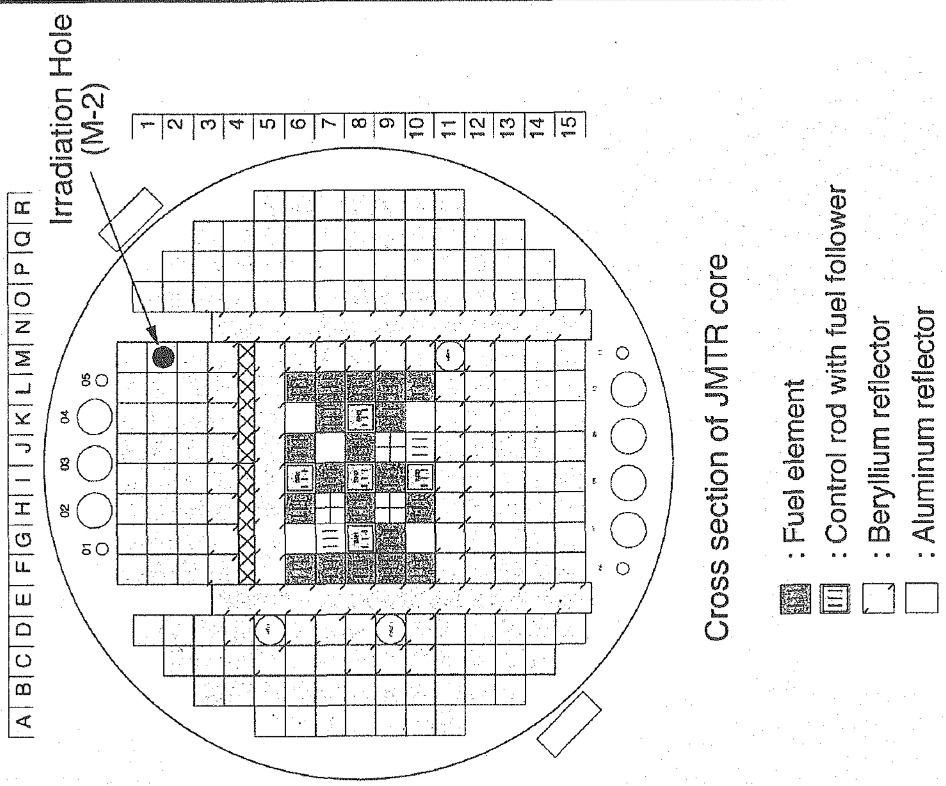


Test Condition

Pebble material : YTZ ($95\%ZrO_2+5\%Y_2O_3$)
Primary pebbles : φ2mm
Secondary pebbles : φ0.1~0.5mm
Container : φ20mmI.D. (Acrylic resin)
Binding force : 32gf/cm^2
Frequency : 48Hz
Amplitude : 1.5mm



Irradiation Position of the Mock-up



Conceptual Structure of the In-pile Mockup and Calculation Model on JMTR Core

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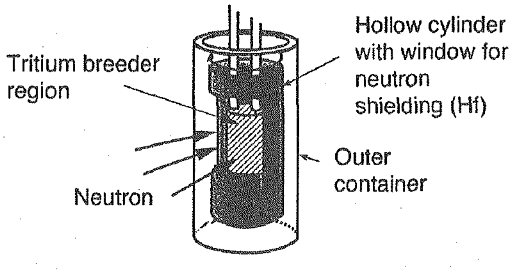
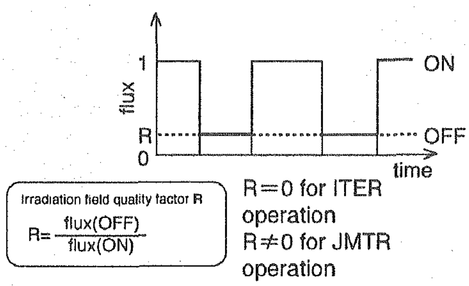
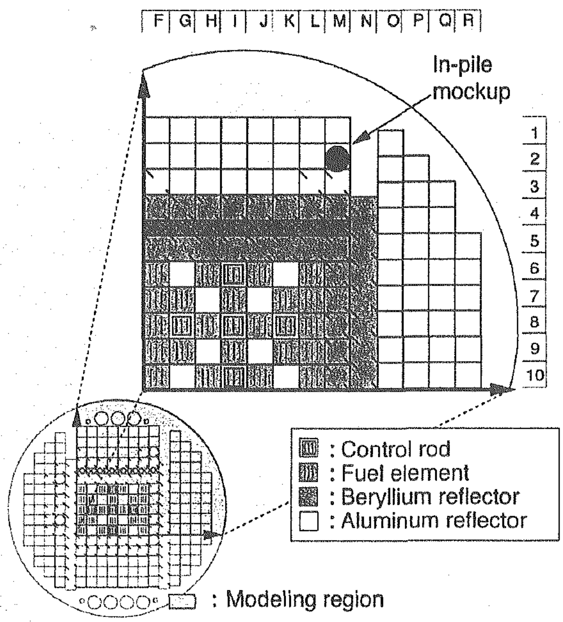


(a) Conceptual structure



(b) Pulse operation mode



Calculation of Thermal Neutron Ratio

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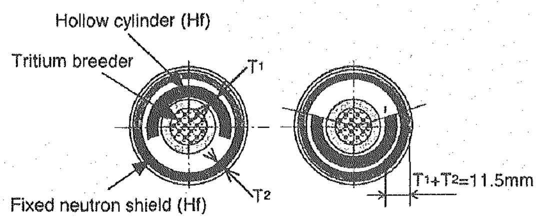
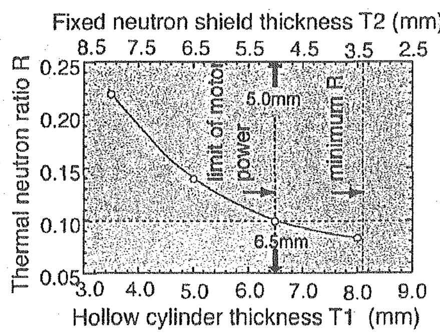
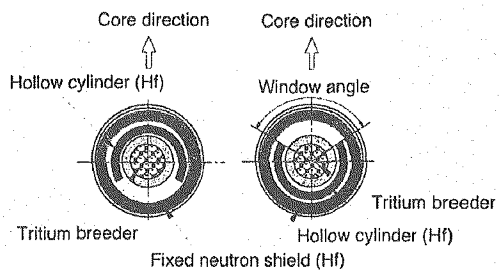
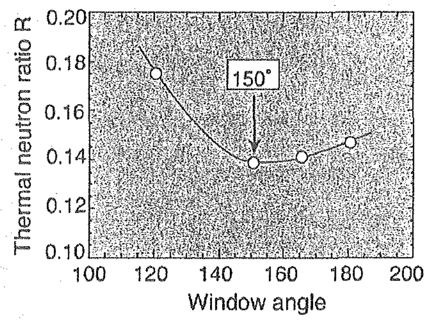
Window angle





Thickness of fixed neutron shield and hollow cylinder





Calculation on Effective Thermal Conductivity of Pebble Bed

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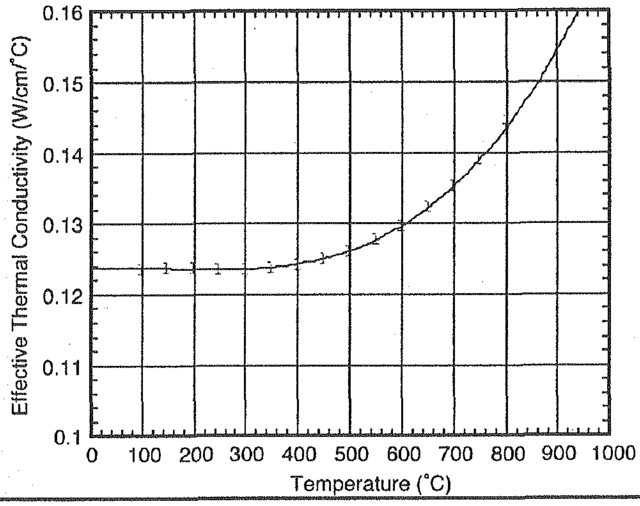
[Cal. Parameter]

- -Packing Fraction : 80%PF (Primary pebble=63%PF, Secondary pebble=17%PF)
- -Li_2TiO_3 pebble diameter : Primary pebble=2mm, Secondary pebble=0.3mm
- -Li_2TiO_3 pebble Density : 83%TD
- -Li_2TiO_3 Thermal Conductivity : Equation by Tsuchiya et al.
- -Effective Thermal Conductivity in Binary Packing : Equation by SZB
  (Schlunder, Zehner and Bauer : SZB)



Calculation Model of Irradiation Temperature in Pebble Bed

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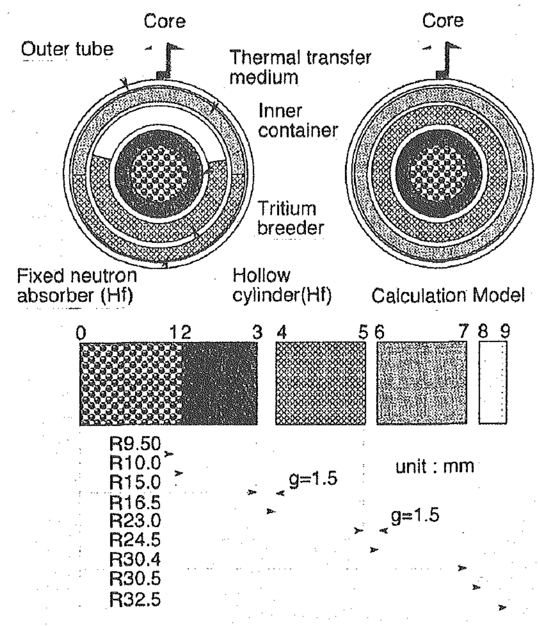
Window Condition : Open　　Window Condition : Close



[Cal. Parameter]

(1) Cooling Condition

- Temperature of cooling water : 50°C
- Heat Transfer Coefficient : 2.33W/cm°C

(2) Property Values

- Lithium Titanate (Li_2TiO_3)
  - a) (Density)=2.28g/cm^3
  - b) Theoretical Density : 3.43g/cm^3
  - c) Sintering Density : 83%TD
  - d) Packing Fraction : 80%P.F
    - Preliminary pebble : 63%F.P
    - Secondary pebble : 17%F.P
  - c) Efficient Thermal Conductivity (W/cm°C)

    $\lambda=1.24\times10^{-2}+2.33\times10^{-7}\cdot T-3.35\times10^{-9}\cdot T^2+7.65\times10^{-12}\cdot T^3$
- Hafnium (Hf)
  - a) Density=13.36g/cm^3
  - b) Thermal Conductivity

    $\lambda=0.230\times10^{0}-7.000\times10^{-5}\cdot T+5.400\times10^{-8}\cdot T^2$ (W/cm°C)
  - c) Thermal Expansion=5.900x10^{-6}(1/°C)

Calculation Results

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Items \ Condition	Window Condition	
	Open	Close
Fast Neutron Fluence (n/cm2/s)	4.9×10^{11}	4.4×10^{11}
Thermal Neutron Fluence (n/cm2/s)	2.0×10^{12}	2.7×10^{11}
Total Neutron Fluence (n/cm2/s)	8.3×10^{12}	5.2×10^{12}
α-heat (W/g)	0.81	0.11
γ-heat (W/g)	0.17	0.14
Tritium Generation (Ci/d)	8.9×10^{-1}	1.3×10^{-1}
Center Temperature of Pebble Bed (˚C)	356	275
Temperature of Edge of Pebble Bed (˚C)	215	264

Result of Tritium Release at Reactor Startup

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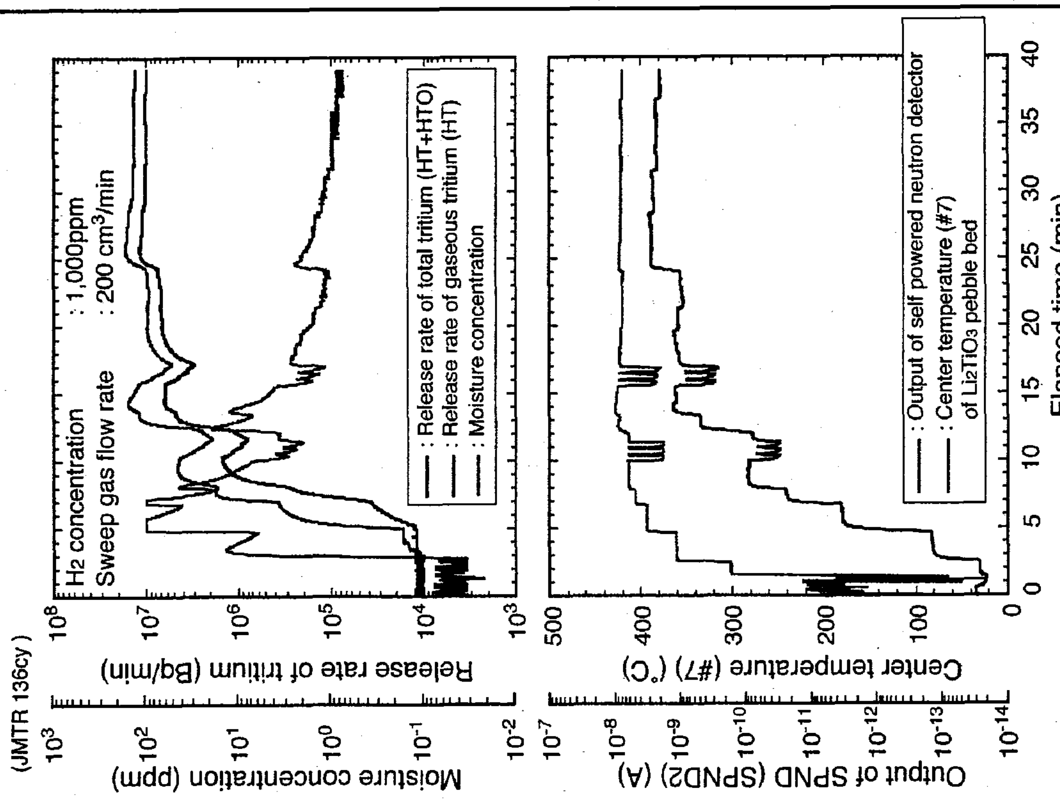
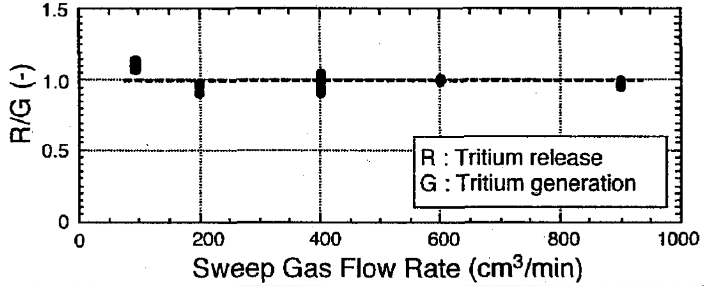


Results of Sweep Gas Flow Rate Changing Tests

JAERI

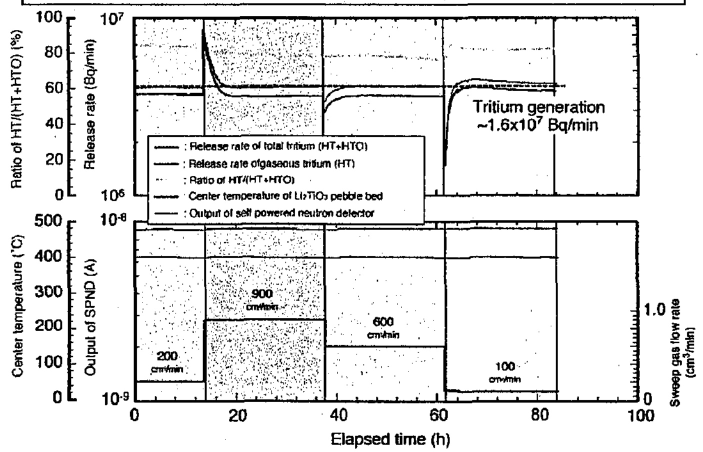
[Results]

1) Amount of tritium recovery was constant at the steady state when sweep gas flow rate changed from 100 to 950 cm³/min (linear flow rate : 0.5~5.0 cm/s).
2) Transients were followed by positive tritium recovery peaks for sweep gas flow rate increases and negative recovery peaks for sweep gas flow rate decreases.



[Experimental conditions]

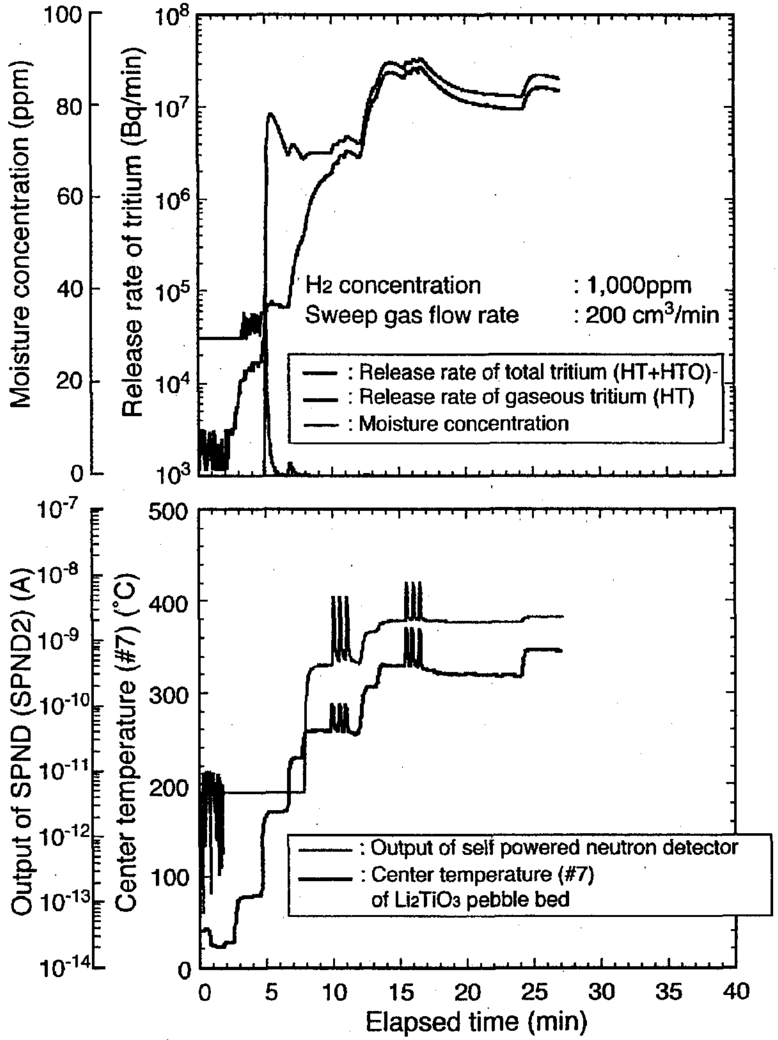
H_2 content	: 1,000 ppm
H_2O concentration	: < 0.3 ppm (in sweep gas)
Irradiation temperature	: ~400°C (center of Li_2TiO_3 pebble bed)
	: ~350°C (edge of Li_2TiO_3 pebble bed)



Result of Tritium Release at Reactor Startup

JAERI

(JMTR 137cy)



Rotating Condition for ITER Nominal Pulsed Operation

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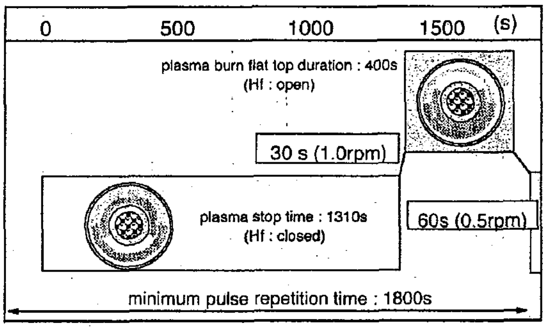
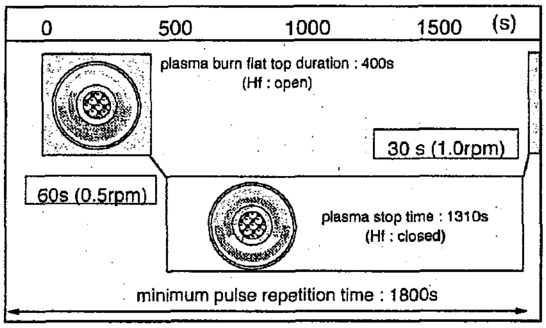
Nominal pulsed operation scenarios
(ITER-FEAT Outline Design Report, G A0 R1 2 99-11-22 W0.1)

Plasma burn flat top duration	400 s
Plasma current flat top length	430 s
Plasma cooling phase	60 s
Plasma current final ramp down	130 s
Minimum pulse repetition time	1800 s

Rotating condition (Run 1) in T431
(closed ⟶ open)



Rotating condition (Run 2) in T431
(open ⟶ closed)



Effect of Irradiation Temperature

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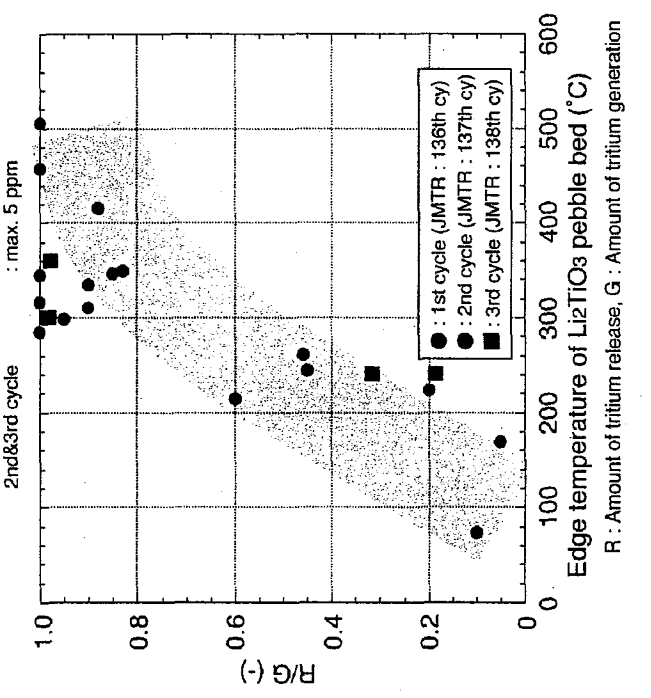
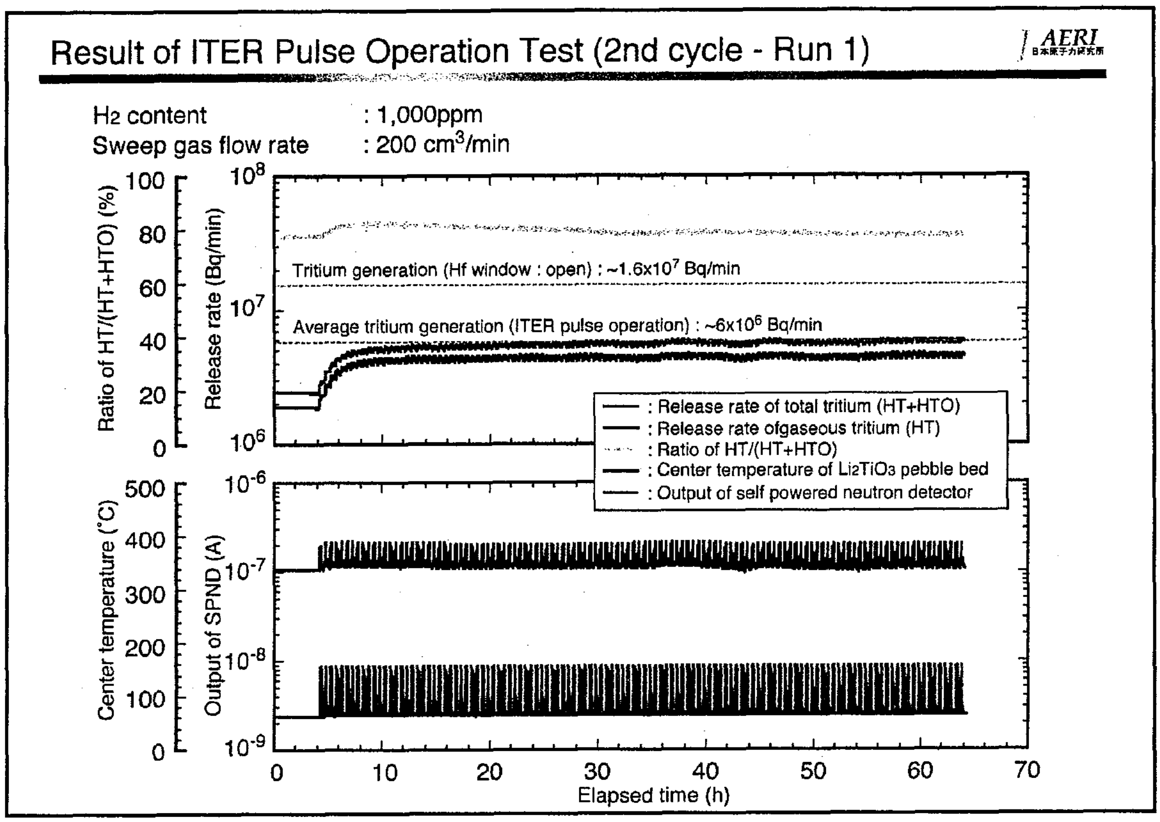
[Results]

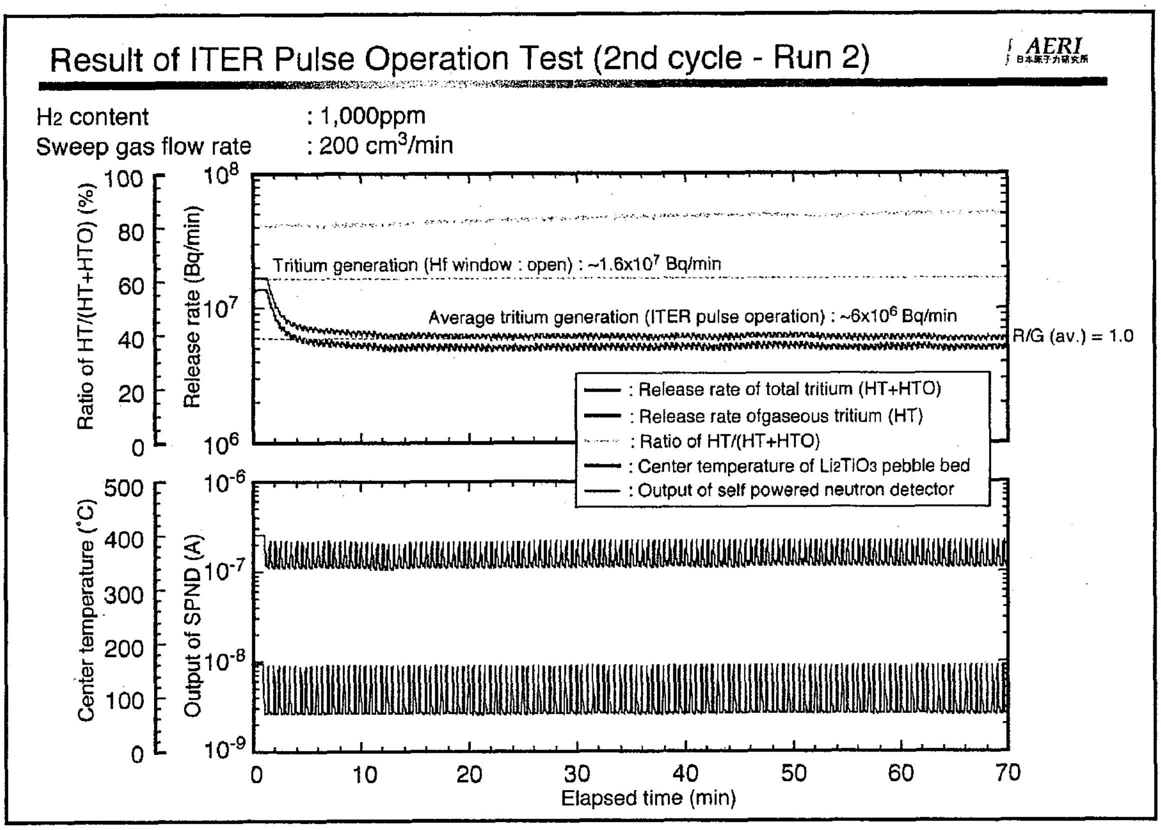
1) R/G increased with increasing irradiation temperature.
2) Tritium release behavior of the first cycle was different of that of second and third cycles. It seems that moisture concentration has an effect on tritium release from Li_2TiO_3 pebble bed.

[Experimental conditions]

Sweep gas flow rate	: 200 cm^3/min
H_2 content	: 1,000 ppm
H_2O concentration	
1st cycle	: 0.3~500 ppm
2nd&3rd cycle	: max. 5 ppm




Result of ITER Pulse Operation Test (2nd cycle - Run 1)
JAERI
H2 content : 1,000ppm
Sweep gas flow rate : 200 cm3/min
Ratio of HT/(HT+HTO) (%)
Release rate (Bq/min)
Tritium generation (Hf window : open) : ~1.6x10^7 Bq/min
Average tritium generation (ITER pulse operation) : ~6x10^6 Bq/min
: Release rate of total tritium (HT+HTO)
: Release rate ofgaseous tritium (HT)
: Ratio of HT/(HT+HTO)
: Center temperature of Li2TiO3 pebble bed
: Output of self powered neutron detector
Center temperature (°C)
Output of SPND (A)
Elapsed time (h)


Result of ITER Pulse Operation Test (2nd cycle - Run 2)
JAERI
H2 content : 1,000ppm
Sweep gas flow rate : 200 cm3/min
Ratio of HT/(HT+HTO) (%)
Release rate (Bq/min)
Tritium generation (Hf window : open) : ~1.6x10^7 Bq/min
Average tritium generation (ITER pulse operation) : ~6x10^6 Bq/min
R/G (av.) = 1.0
: Release rate of total tritium (HT+HTO)
: Release rate ofgaseous tritium (HT)
: Ratio of HT/(HT+HTO)
: Center temperature of Li2TiO3 pebble bed
: Output of self powered neutron detector
Center temperature (°C)
Output of SPND (A)
Elapsed time (min)