Fusion Engineering and Design 146 (2019) 1077–1081

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

Li-rod structure in high-temperature gas-cooled reactor as a tritium production device for fusion reactors

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ARTICLE INFO

Keywords:
Tritium
Lithium
Zirconium
Lithium-loading rod
High-temperature gas-cooled reactor
HTTR

ABSTRACT

Production of tritium using a high-temperature gas-cooled reactor (HTGR) has been studied for a prior engineering test with tritium handling and for the startup operation of a demonstration fusion reactor. For this purpose, the hydrogen absorption speed of Zr in a Li-loading rod for the reactor operation is experimentally measured, and an analysis model is presented to evaluate the tritium outflow from the Li rod in a high-temperature engineering test reactor (HTTR). On the basis of the presented model, the structure of the Li-loading rod for the demonstration test using the HTTR is proposed.

1. Introduction

For prior technical tests of tritium circulation and blanket systems of a demonstration fusion reactor and for the startup operation, it is necessary to provide a sufficient amount of tritium from an outside source [1]. For fusion reactors, the tritium has been produced by using the D(n,γ)T reaction in the Canadian Deuterium Uranium (CANDU) reactors [2]. Because the cross section of the ^{6}Li(n,α)T reaction has almost 6 orders larger around the thermal-neutron energy range compared with the D(n,γ)T reaction and the high-temperature gas-cooled reactor (HTGR) has the following several advantages, we consider the tritium production using the HTGR [3] by inserting a Li compound as a burnable poison (BP) instead of a boron compound. The main core structure in the HTGR is graphite, which is chemically stable and does not react with the Li compound. The large core size of the HTGR provides enough space for loading the Li compound without ^{6}Li enrichment, together with structural materials to prevent the leakage of tritium. In the HTGR, the BP is usually used in a solid state (i.e., as B_4C), and thus the Li compound can be loaded into the reactor's core without significantly changing the original structural design. The technical understanding of the Li-loading rod structure, together with the evaluation of the basic properties of the structural materials of the rod (i.e., Al_2O_3 and Zr [4–8]), has been progressed to efficiently produce a sufficient amount of tritium and to hold the produced tritium inside of the rod during the reactor's operation period. In order to confirm the performance of the Li-loading rod and to demonstrate the tritium production process using the HTGR, we now plan an irradiation test on a high-temperature engineering test reactor (HTTR). HTTR is a block-type high-temperature gas-cooled fission reactor with a 30-MW thermal output [9], which is operated by the Japan Atomic Energy Agency. Heading on the irradiation test, the main structure of the Li-loading rod with a capsule for tritium containment should be clarified.

During the operation, the amount of tritium flowing out from the Li rod into helium gas should be suppressed to a lower level as much as possible. Numerical simulations have predicted that if we could operate the HTGR in a low temperature range, keeping the rod temperature below 520 °C, the tritium leaking from the Li rod can be suppressed to less than 1% of the amount produced [4]. However, if we intended to operate the HTGR in a much higher temperature range (i.e., the rod temperature reaching 800 °C–900 °C) so as to increase the electricity generation efficiency, the leakage of the tritium would rapidly increase. This is because the hydrogen permeability of the Al_2O_3 layer in the Li rod increases with the increase of the rod's temperature [5]. In order to reduce the leakage of tritium from the rod, we attempted to adopt Zr in the rod as a tritium absorption material to avoid increasing the inner tritium partial pressure [6,8,10]. To prevent the tritium absorption

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https://doi.org/10.1016/j.fusengdes.2019.02.009
Received 12 September 2018; Received in revised form 4 February 2019; Accepted 4 February 2019
Available online 08 March 2019

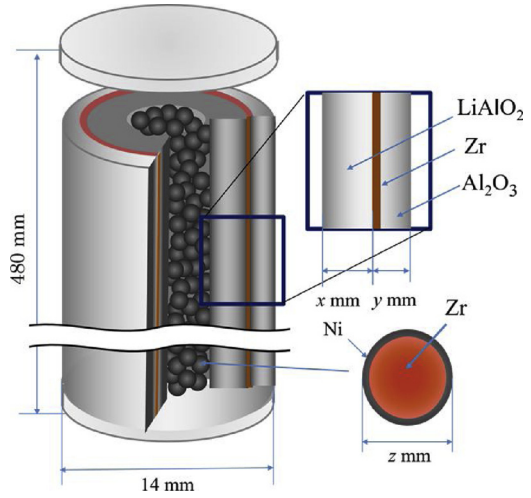

Fig. 1. A schematic view of the Li-loading rod.

capability from decreasing due to oxidization via interacting with co-existing oxides (i.e., $LiAlO_2$ and Al_2O_3), some kind of coating with an antioxidant material is necessary [10]; it is desirable to have a surface area of Zr is as large as possible. We propose the Li rod including Zr pebbles with Ni coating as shown in Fig. 1.

The optimal size of each layer and the diameter of the pebble (e.g., *x*, *y*, and *z* in Fig. 1) should be clarified. In this paper, we experimentally measured the hydrogen absorption performance of Zr (i.e., hydrogen absorption speed). On the basis of the measured data, we numerically simulated the tritium leakage from the Li rod during the reactor's operation and the correlation with the rod structure, assuming several patterns of the tritium absorption capability. The purpose of this paper was to present a desirable Li-rod structure and its analysis model for tritium production in the HTTR.

2. Experimental apparatus and analysis model

2.1. Hydrogen absorption experiment

Hydrogen absorption experiments were carried out using two commercial Zr cylinders (see Table 1). A schematic illustration of the experimental apparatus is shown in Fig. 2. A Zr cylinder is introduced in the center region of a quartz tube, which is heated to the preset temperature (i.e., 850 °C or 900 °C); this temperature is kept constant during the measurement. After the inside of the quartz tube is evacuated, hydrogen gas flows into the tube so that the initial pressure becomes ~500 Pa. The pressure reduction process in the quartz tube due to hydrogen absorption by Zr is continuously observed.

Table 1
Properties of Zr cylinders.

	Small	Large
Diameter (mm)	9.5	15.8
Height (mm)	15	15
Thickness (mm)	1	1
Impurity		
C (wt%)	0.015	
FeCr (wt%)	0.083	
H (wt%)	< 0.0003	
N (wt%)	0.006	
O (wt%)	0.143	
Zr + Hf (wt%)	99.5	

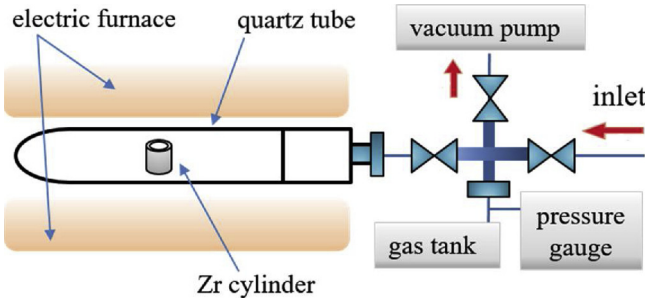

Fig. 2. Illustration of the experimental apparatus.

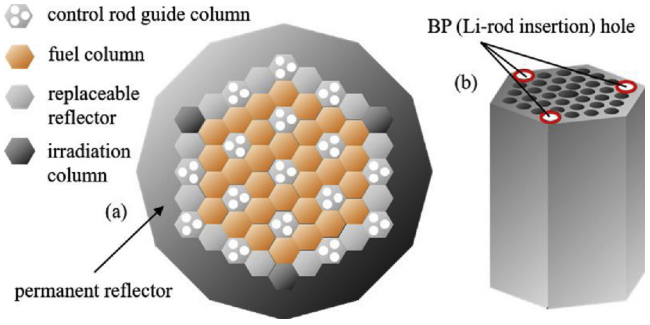

Fig. 3. (a) Horizontal cross-section of the HTTR core, (b) Fuel block with Li-rod insertion holes.

2.2. Nuclear burning simulation for HTTR core

The horizontal cross section of the HTTR core is shown in Fig. 3 (a) [9]. The core consists of 2 regions, i.e., actual core and reflector regions. The actual core consists of 30 fuel and 7 control-rod guide columns, each of which is composed of a stack of 5 fuel and 4 reflector blocks. The actual core is surrounded by the reflector region, i.e., replaceable and permanent reflector blocks and 9 control-rod guide blocks. The core is 2.9 m in height and 2.3 m in diameter. Each hexagonal block is 360 mm wide across the flats and 580 mm high [see Fig. 3 (b)]. The fuel block contains 31 or 33 fuel channels and 3 BP insertion holes. 480 mm high Li rods with 15 mm Al_2O_3 caps on both upper and bottom sides are loaded into the BP insertion holes.

The estimation of the amount of produced tritium and the effective multiplication factor during the HTTR operation was performed using the continuous-energy Monte Carlo transport code MVP-BURN [11] using the whole core model. Throughout the calculations, nuclear data were taken from JENDL-4.0 [12]. The fuel region temperature was 1100 °C and the Li-rod temperature was the same as the moderator (900 °C). In this study we assumed 360-day reactor operation period. All control rods were assumed to be pulled out. The time steps in the burn-up simulations were taken as 0, 1, 5, 30, 60, 120, 180 and 360 days. For each of the time steps, 6,000,000 neutrons were generated. The statistical errors of the effective multiplication factor and the reaction rate were less than 0.1% in all calculations, which is sufficient accuracy in our discussion.

2.3. Diffusion equation and outflow of tritium

The total amount of tritium that flowed out from the Li rods into the helium coolant per unit time was estimated by solving the tritium diffusion equation. We ignored the function of Zr as an antipermeation material and solved the diffusion equation only for the Al_2O_3 layer:

$$\frac{\partial c}{\partial t} = D\nabla^2 c, \tag{1}$$

where *D* and c represent the diffusivity of the hydrogen isotope in Al_2O_3

and the tritium density in the hollow portion, respectively. This treatment can estimate the tritium leakage on the safe side. Tritium produced in the $LiAlO_2$ layer diffuses quickly in the hollow portion, and the pressure of tritium in the $LiAlO_2$ and the hollow portion layers reach the same value. It has already been confirmed that hydrogen permeates through the Al_2O_3 tube in an atomic form (i.e., hydrogen concentration is proportional to a square root of an input hydrogen pressure [5]), thus we determined the boundary condition of Eq. (1) at the inner surface of the Al_2O_3 tube on the basis of Sieverts' law. The outflow of tritium from the outer surface of the Al_2O_3 tube into the coolant was estimated from a nonequilibrium solution of the tritium diffusion equation assuming the one-dimensional cylindrical geometry as

$$J = -AD\frac{\partial c}{\partial r}\bigg|_{r=7\text{mm}}, \tag{2}$$

where A is the outer surface area of the Al_2O_3 tube. The diffusivity and solubility coefficients of Al_2O_3 for a hydrogen isotope were taken from the work of Katayama [5]. The porosities of Al_2O_3 and $LiAlO_2$ are assumed to be 0 and 15% respectively.

3. Results and discussion

3.1. Tritium production in the HTTR

The effective multiplication factor (k_{eff}) and the cumulative weight of tritium produced in the HTTR are plotted in Fig. 4 as a function of operation time for several $LiAlO_2$ thicknesses. Here the effective multiplication factor represents how the reactor has allowable margin for the neutron flux to sustain the criticality at the indicated operation time, and the simulation is made assuming the core in which all control rods are pulled out. In the actual operation, the k_{eff} is kept unity by using the control rods during the operation. In the simulations, a 14 mm diameter of the Li-rod, a 1.9 mm thickness of the Al_2O_3 layer, and a 0.1 mm thickness of the Zr layer are assumed, implying that the thickness of the $LiAlO_2$ layer in Fig. 4 was changed by adjusting the inner radius of the $LiAlO_2$ layer, keeping the outer radius as 5 mm. k_{eff} (the cumulative weight of the produced tritium) kept decreasing (increasing) over time and reached the minimum (maximum) value after the 360-day operation.

The cumulative amount of produced tritium and the effective multiplication factor right after the 360-day operation was terminated are summarized in Fig. 5 as a function of the $LiAlO_2$ thickness. Although the amount of produced tritium increases for large $LiAlO_2$ mass, k_{eff} decreases. In order to continue operating the reactor, at least 1.02 of k_{eff} is required until the end of the operation. From Fig. 5 we can estimate that the possible weight of tritium produced in the HTTR is roughly 30 g during 360-day operation. Zr was not included in the simulation; however, the weight of Zr is not a crucial factor for tritium production. This is because the neutron absorption by Zr is small. In the following discussion we will use the value of 30 g/year as the typical weight of tritium produced in the HTTR.

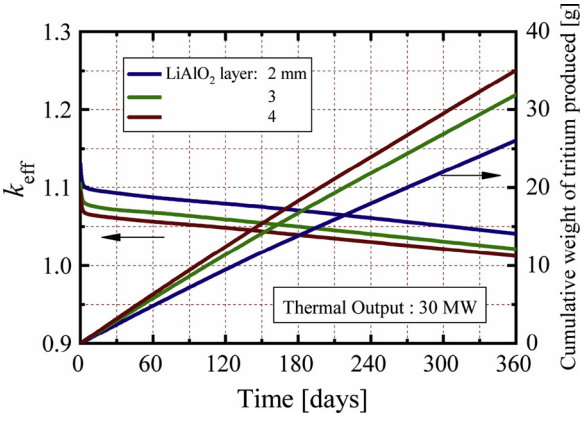

Fig. 4. The effective multiplication factor (k_{eff}) and the amount of tritium produced for each operation time.

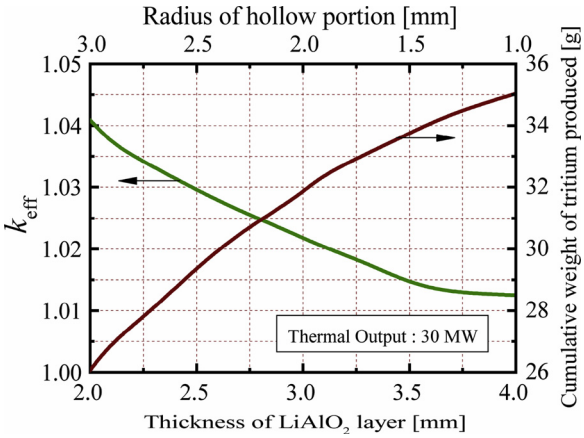

Fig. 5. Effective multiplication factor (k_{eff}) and the amount of tritium produced during 360-day operation in the HTTR.

3.2. Estimation of hydrogen absorption time

In order to estimate the tritium leakage from the Li rod in which Zr is inserted as a hydrogen absorption material, the absorption speed and the quantity possibly absorbed should be considered. Hydrogen absorption experiments were carried out to examine the hydrogen absorption speed and its quantity. The results of the experiments (i.e., temporal behaviors of the hydrogen pressure due to absorption by the Zr cylinders) are shown in Fig. 6. The dotted lines represent the case when the Zr temperature was kept at 850 °C, and the solid lines represent the case when the Zr temperature was kept at 900 °C. Since the absorption speed is predicted to be related to the size of the surface area of the Zr sample, we prepared samples of small and large sizes as shown in Table 1. The black triangles show the samples with a small size, whereas the white squares represent the samples with a large size. We can find that the absorption of hydrogen progresses according to the exponential scaling (dotted lines show the exponential fittings), and we can estimate the absorption time (τ_a) by least squire fitting as follows:

$$\hat{P} \approx \exp(-t/\tau_a) + \hat{P}_{equilibrium}. \tag{3}$$

Here $\hat{P}_{equilibrium}$ is the normalized pressure when $t\to\infty$, which is sufficiently small compared with $\hat{P}$. We define τ^*_a using a product of τ_a and Zr surface area:

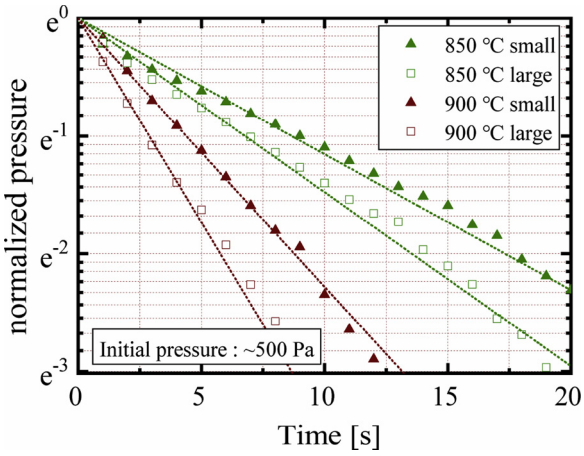

Fig. 6. Temporal behaviors of normalized hydrogen pressure.

$$\tau_a^* \equiv \tau_a^{small} A^{small}/V_{ex} \approx \tau_a^{large} A^{large}/V_{ex}, \quad (4)$$

where A^{small} (A^{large}) is the surface area of small (large) sized sample and V_{ex} is the volume filled with the hydrogen gas. In our experiment, τ_a^{small} = 8.7 (4.4) s and τ_a^{large} = 6.8 (2.9) s were observed, and $\tau_a^* \approx$ ~9.4 (4.3) s/m for 850 (900)°C is almost of the same value for both samples, which implies that the absorption speed is approximately proportional to size of the surface area. In our experiment the solubility of the Zr is measured as 91.3 (75.5) mol/m^3/Pa$^{1/2}$ for 850 (900)°C.

3.3. Evaluation of tritium outflow and rod parameters

We next evaluated the amount of tritium outflow from the rods by solving Eq. (1). In order to determine the boundary condition at the inner surface of the Al_2O_3 layer (according to Sieverts' law), it is necessary to know the tritium partial pressure at the surface. We assume that the tritium density in the hollow portion satisfies the following mass balance equation:

$$\frac{dc}{dt} = \frac{S_T^{rod}}{V_{hp+Li(15)}} - \frac{cA_{pebble}}{\tau_a^* V_{hp+Li(15)}} + \frac{AD}{V_{hp+Li(15)}} \frac{\partial c}{\partial r}\bigg|_{r=7\text{mm}}, \quad (5)$$

where S_T^{rod} is the tritium production rate, and A_{pebble} represents the total surface area of the Zr pebbles, and $V_{hp+Li(15)}$ is the volume of 15% $LiAlO_2$ layer plus hollow portion. From the solution of Eq. (5), the partial pressure of tritium in the hollow portion (see Fig. 1) was estimated using the equation of state for a perfect gas.

In the HTGR, a large core size makes it possible to load a large amount of Zr in the Li rod. The atomic ratio T/Zr can be estimated to be less than 0.1 at most, even after the 360-day operation. In such a case, as shown by Zuzek [13], the capability of tritium absorption in Zr would be sufficiently high to reduce the partial tritium pressure, as long as the ideal condition is kept satisfied during the operation. Yamanaka [14] reported that the relative reduction of the solubility due to oxidization is restricted to 1/3 ~ 1/2 in the HTGR condition when O/Zr atomic ratio is within 0.25 ~ 0.398; thus in the HTGR we could roughly know the influence of the Zr oxidization on the tritium leakage by looking at the decrement of the absorption speed relative to the tritium production.

The weight of tritium flowing out from all rods for 900 °C and the inner pressure are presented in Fig. 7 as a function of the thickness of the Al_2O_3 layers for several τ_a^{rod}, i.e., τ_a^{rod} = 1 h, 1 day, 10 days, 60 days and ∞ for the weight of tritium flowing out (solid lines) and τ_a^{rod} = 60 days and ∞ for the inner pressure (dotted lines). Here τ_a^{rod} is the τ_a when thickness of the Al_2O_3 layer is 1.9 mm. In the calculations, the thicknesses of the both Zr layers inside and outside of $LiAlO_2$ surface are assumed to be 0.1 mm. The thickness of the $LiAlO_2$ layer is adjusted so that total tritium production can be kept at 30 g. We simulate the

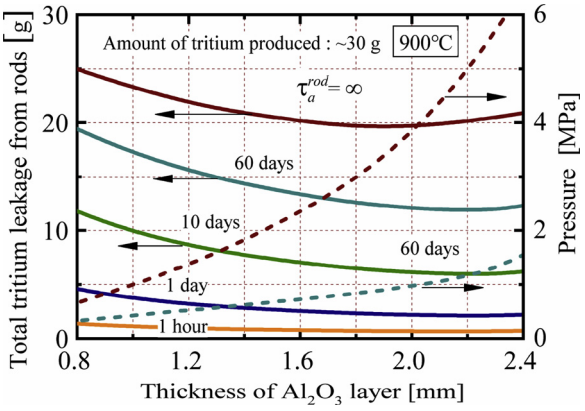

Fig. 7. Weight of tritium leakage from the rods (solid lines) and inner pressure in one rod (dotted lines) as a function of the Al_2O_3 layer thickness for several τ_a^{rod} when 30 g of tritium is produced.

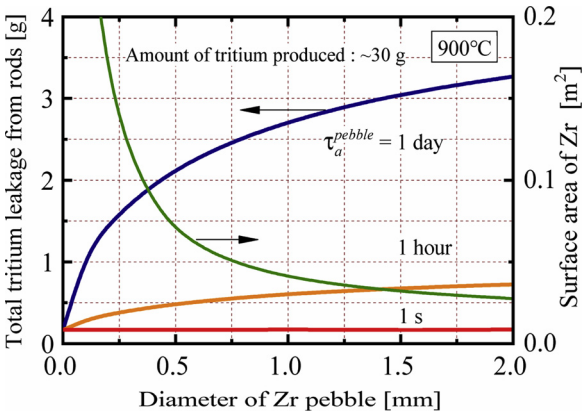

Fig. 8. Weight of tritium leakage from the rods as a function of the Zr pebble diameter for several τ_a^{pebble} when 30 g of tritium is produced.

reduction of the tritium absorption capability of Zr due to some reason as an increment of the absorption time τ_a. It is found that with increasing the absorption time, the fraction of tritium leakage to the amount of production increases. The condition $\tau_a^{rod} = \infty$ corresponds to the case when the rod without Zr is adopted. We can understand that if the thickness of Al_2O_3 layer is less (greater) than 1.8 ~ 2.2 mm, tritium leakage decreases (increases) with the increase of the thickness. This is because the inner pressure increases owing to the decrement of the volume of the hollow portion. Under this condition, almost 2/3 of the produced tritium flows out if we do not use Zr. In our experiment, τ_a is estimated as 1.2 ms in the rod. If the performance of Zr absorption works well as measured in our experiment, or at least τ_a is shorter than 1 day, the leakage can be reduced to less than ~1.0 g. The difference in ~10^8 (between ms and day) roughly corresponds to the difference of the diffusion coefficient for hydrogen between Zr and ZrO_2.

In Fig. 8 the ratio between the tritium leakage and the amount of produced tritium is plotted as a function of the diameter of the Zr pebble for some τ_a. In the simulations, a 14 mm diameter of the Li-rod, a 1.9 mm thick Al_2O_3 layer, a 0.1 mm thick Zr layer, and a 2.7 mm thick $LiAlO_2$ layer are assumed. In this case, the diameter of the hollow portion becomes 4.6 mm and the amount of tritium produced during the 360-day operation is 30.6 g. The volume ratio of the Zr pebbles inside the $LiAlO_2$ layer is fixed as 60%, and we define τ_a^{pebble} as the τ^a when diameter of Zr pebble is 1.0 mm. As the diameter of Zr pebble decreases, the total surface area of the Zr increases, and the amount of tritium leakage decreases. The decrement of the diameter effectively works to reduce the tritium leakage when τ_a > 1 day. When τ_a is shorter than one hour, we can say that the leakage becomes nearly the same level as the one generating in usual HTTR operation without Li loading, i.e., ~0.25 g/year [15].

4. Concluding remarks

In this paper, we described an analysis model for evaluating tritium leakage from a Li-loading rod and proposed a desirable rod structure for tritium production in an HTTR. At this point, we consider that a rod with a 14 mm diameter, a 1.9 mm thick Al_2O_3 layer, a 0.1 mm thick Zr layer, and a 2.7 mm thick $LiAlO_2$ layer, involving Zr pebbles with Ni coating and a 0.5 mm radius, is one of the most desirable structures of a Li-rod for an HTTR.

In fission reactors, all of the materials are exposed to intensive radiation of neutrons. In such a case the hydrogen diffusion may be retarded due to the trapping effect at the radiation defects [16]. On the other hand enhancement of hydrogen diffusion by radiation for oxide including Al_2O_3 was reported [17]. The oxides can be potential candidates of permeation barrier, and the radiation-induced diffusion may deteriorate the barrier effect [18]. At this point, the mechanism of the

deterioration has not been understood fully. In order to complete the final design, the amount of tritium produced and leaked (i.e., the analysis model and the effective hydrogen absorption time of Zr) should be validated in an irradiation test in an HTTR.

Acknowledgments

This work is supported by JSPS KAKENHI Grant-in-Aid for Scientific Research (B) JP18H1200 and cooperative program with BA (The Joint Implementation of the Broader Approach Activities in the Field of Fusion Energy Research) reactor design team in Japan.

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