

Evaluation of hydrogen permeation rate through zirconium pipe

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ABSTRACT

To launch a fusion reactor stably, it is necessary to prepare sufficient amount of tritium by an external tritium source. Tritium production using nuclear reactions by neutron and Li in a high temperature gas-cooled reactor (HTGR) is an attractive method. An important issue is tritium confinement in high temperature conditions of HTGR. Covering Li compound by Al_2O_3 is a promising method because hydrogen permeability in Al_2O_3 is quite low. Furthermore, it is expected that inserting Zr between Li compound and Al_2O_3 suppresses tritium permeation because Zr has a large hydrogen absorption capacity. For example, a cylindrical multilayer structure consisted of Zr – Li compound – Zr – Al_2O_3 is a candidate structure to confine generated tritium. To evaluate hydrogen permeation rate through cylindrical Zr material, in this work, hydrogen permeation experiments were carried out for two samples of one-side sealed Zr pipe. Hydrogen permeation rate in one sample was proportional to as the square root of hydrogen pressure but that in another sample, which had retained a certain amount of hydrogen before the experiment, did not indicate an obvious pressure dependence regardless of almost the same experimental procedure except hydrogen permeation direction. Since observed hydrogen permeation rate in Zr was faster than that in Al_2O_3 , under the multi confinement structure by Zr – Li compound – Zr – Al_2O_3 , the generated tritium in Li compound diffuses promptly in whole Zr inside Al_2O_3 layer and it is expected tritium is stored stably in the structure.

1. Introduction

Preparation of sufficient tritium by an external tritium source is necessary to launch a fusion reactor stably. Tritium production using nuclear reactions by neutron and Li in a high temperature gas-cooled reactor (HTGR) is an attractive method [1]. An important issue is tritium confinement in high temperature conditions of HTGR. Covering Li compound by ceramics coating such as Al_2O_3 and PyC is a promising method because hydrogen permeability in these material is quite low [2,3]. Furthermore, it is expected that incorporating Zr between Li compound layer and a ceramic coating layer suppresses tritium permeation because Zr has a large hydrogen absorption capacity. Katayama et al. indicated tritium confinement performance of an Al_2O_3 -coated Li compound particle containing Zr fine particles by the numerical simulation using literature data of hydrogen behavior in Zr [2]. Nakaya et al. proposed the Li rod consist of cylindrical LiAlO_2 coated by Zr and Al_2O_3 [4]. Since the Li rod has a small surface area as compared with a large number of Li particles of the same mass of Li, total tritium permeation rate from the same mass of Li becomes small. Additionally, by sandwiching the inside and the outside of cylindrical LiAlO_2 with Zr layer, tritium produced in LiAlO_2 can be effectively recovered by Zr.

The present authors have a plan to test tritium confinement performance of Zr layer – Li compound – Zr layer – Al_2O_3 layer structure by input tritium gas instead of Li compound. As a first step of the study, understanding of hydrogen permeation behavior through Zr layer is important. However, experimental data for hydrogen permeation through Zr material is few. In many cases, hydrogen solubility or hydrogen diffusivity or both of that were separately reported. Therefore, in this work, hydrogen permeation behavior through one-side sealed Zr pipes was observed at moderate temperature and the permeation rate was evaluated.

2. Experimental

Two samples of one-side sealed Zr pipe were used in hydrogen permeation experiment. One is 9.5 mm in outer diameter, 8.0 mm in inner diameter, 210 mm in length and 1 mm in bottom thickness. Another one is 6.3 mm in outer diameter, 4.0 mm in inner diameter, 200 mm in length and 1 mm in bottom thickness. The sample pipes were made by drilling out the center of the round bar of Zr by a metal processing company. Each Zr pipe was joined with a stainless steel pipe by a heat resistant resin.

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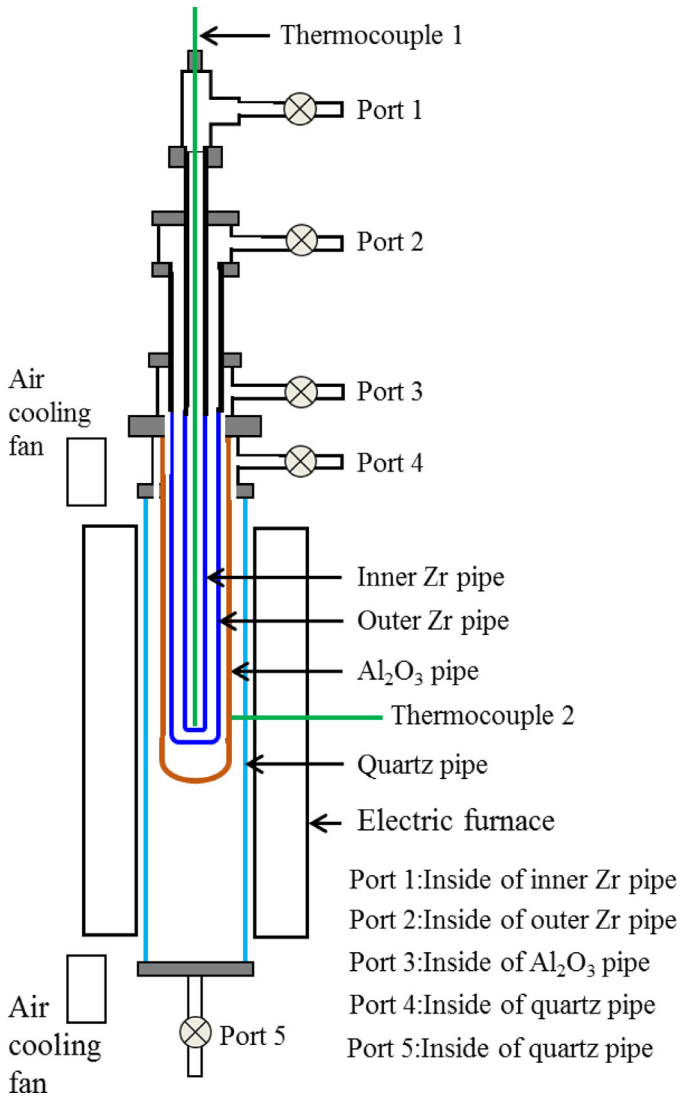


Fig. 1. Schematic of the experimental apparatus.

As mentioned in the previous section, the authors plan to test the tritium confinement performance of the multilayer structure of Zr–Zr–Al₂O₃ in the near future. The permeation experiments of hydrogen through Zr pipe were conducted by using the apparatus to be used in the test. The schematic illustration of the experimental apparatus is shown in Fig. 1. The apparatus has a quadruple pipe structure of two one-side sealed Zr pipes, a one-side sealed Al₂O₃ pipe and a quartz pipe. The Al₂O₃ pipe was inserted into the quartz pipe. And the outer Zr pipe was inserted into the Al₂O₃ pipe, finally the inner Zr pipe was inserted into the outer Zr pipe.

The hydrogen permeation experiments through the outer Zr pipe and the inner Zr pipe were conducted separately. When the experiment for the outer Zr pipe is performed, the inner Zr pipe was replaced to a stainless steel pipe that both side is not sealed. The mixed gas of hydrogen and argon was introduced to the port 1 and passed through the inside of the outer Zr pipe and released from the port 2 to the atmosphere. The space between the outer Zr pipe and the Al₂O₃ pipe were vacuumed from the port 3 and the permeated hydrogen was measured by a quadrupole mass spectrometer (QMS). When the experiment for the inner Zr pipe is performed, the outer Zr pipe was replaced to a stainless steel pipe that both side is not sealed. The mixed gas of hydrogen and argon was introduced to the port 3 and passed through the outside of the inner Zr pipe and released from the port 2 to the atmosphere. The space inside the inner Zr pipe were vacuumed from the port

1 and the permeated hydrogen was measured. The lower part of the pipes was heated by an electric furnace and the outside of the furnace was cooled by air cooling fans. The furnace temperature was controlled to be the preset temperature by the thermocouple 2, which was contacted on the outer surface of the central portion of the quartz pipe. A one-side sealed quartz pipe, 2.7 mm in outer diameter, was inserted into the center of multi pipes and the thermocouple 1 was inserted into it. Thermocouple 1 was slid up and down inside the quartz pipe to measure the temperature distribution.

In order to investigate the pressure dependence of hydrogen permeation, 2% H₂/Ar gas and 20% H₂/Ar gas were used as sample gases, where hydrogen pressure corresponds to 2027 Pa and 20,270 Pa under atmospheric pressure, respectively. The flow rate was controlled to be 100 sccm by a mass flow controller. The hydrogen supply side is referred to as the primary side and the vacuum side is referred to as the secondary side. During elevating temperature to 500 °C, pure Ar gas was flowed in the primary side and the secondary side was vacuumed. After the temperature was kept to be 500 °C for several hours, the Ar gas was switched to the mixed gas of hydrogen and argon. After the current of hydrogen permeated was stable, the temperature was elevated to the next temperature.

When a diffusion process of hydrogen in a material is the rate controlling step of permeation, the permeation rate in steady state condition, Q [mol/s] is derived from Fick's law and expressed as follows [5],

$$Q = D(C_p - C_s)A/t, \quad (1)$$

where D is the diffusivity [m²/s], C_p and C_s are hydrogen concentration in the top surface layer of the material in the primary side and the secondary side [mol/m³], A is the effective surface area [m²], t is the thickness of the material [m].

In this experiment, since the secondary side was vacuumed, C_s can be set to be zero. Additionally, because temperature distribution was formed in Zr pipe, the permeation rate at each position in the length direction has to be considered. When dividing Zr pipe into n segments, the permeation rate through the side wall in i th segment, $Q_{sw,i}$ is expressed by the following equation,

$$Q_{sw,i} = D_{sw,i}(C_{p,sw,i} - 0)A_{sw,i}/t_{sw}. \quad (2)$$

The effective permeation area of the side wall in i th segment is given by,

$$A_{sw,i} = 2\pi l_{sw,i} t_{sw} / \ln(r_2/r_1), \quad (3)$$

where, l_i is the pipe length in i th segment [m], r_1 and r_2 are inner and outer radii of Zr pipe [m]. The permeation flux through the side wall in i th segment, $J_{sw,i}$ [mol/m²/s] is given by

$$J_{sw,i} = Q_{sw,i}/A_{sw,i}. \quad (4)$$

The permeation rate through the bottom wall, Q_{BW} is expressed by the following equation,

$$Q_{BW} = D_{BW}(C_{p,BW} - 0)A_{BW}/t_{BW}. \quad (5)$$

The effective permeation area of the bottom wall is assumed as follows,

$$A_{BW} = \pi r_1^2. \quad (6)$$

The permeation flux through the bottom wall, J_{BW} [mol/m²/s] is given by

$$J_{BW} = Q_{BW}/A_{BW}. \quad (7)$$

The total permeation rate through the whole Zr pipe, Q_{total} is given by,

$$Q_{total} = \sum_{i=1}^n Q_{sw,i} + Q_{BW}, \quad (8)$$

From Eqs (1) and (4), the following equation is obtained,

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