

# Fabrication of the Hydrogen Recovery Unit in the Molten Salt Loop Orosh<sup>2</sup>i-1 and Preliminary Evaluation for Hydrogen Transfer<sup>\*</sup>)

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Hydrogen recovery unit is developed for the molten salt loop Orosh<sup>2</sup>i-1. Pure Ni was selected as hydrogen permeation material due to its industrial maturity of fabrication technology and good compatibility with molten salt in fusion reactor condition. No significant degradation of hydrogen permeability of the pure Ni during the fabrication process. Advanced hydrogen permeation materials, such as Pd, V, Nb, and Ta, maintaining higher hydrogen permeability are also discussed to develop more compact hydrogen recovery systems.

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## 1. Introduction

In order to demonstrate the compatibility between fuel tritium recovery and heat recovery for fusion reactor system with molten salt breeder blanket, an experiment with a molten salt loop, Operational Recovery Of Separated Hydrogen and Heat Inquiry - 1 (Orosh<sup>2</sup>i-1), has been initiated in National Institute for Fusion Science [1]. The Orosh<sup>2</sup>i-1 loop is going to simulate circulation of molten salt containing tritium among fusion blanket, tritium recovery system and heat exchanger. Hydrogen gas and Flinak (LiF + NaF + KF) are used as stimulants for tritium and Flibe (LiF + BeF<sub>2</sub>) in the Orosh<sup>2</sup>i-1 loop.

One of proposed mechanisms for hydrogen recovery is permeation through metal wall [2]. The candidate hydrogen permeation materials are Ni, Pd, V, Nb, Ta, and their alloys [2–6]. Pure Ni is the most feasible due to its industrial maturity for fabrication, good compatibility with the molten salt, and lower cost than the other candidates. In the present study, hydrogen recovery unit was fabricated with pure Ni and then was evaluated for hydrogen recovery from He-H<sub>2</sub> gas mixture prior to installation to Orosh<sup>2</sup>i-1 loop. Based on the hydrogen permeability for the pure Ni

and also other candidates, feasibility of hydrogen recovery unit for fusion reactor is discussed.

## 2. Fabrication of the Hydrogen Recovery Unit

Figure 1 is the conceptual design for the hydrogen recovery unit for Orosh<sup>2</sup>i-1 loop. The unit is connected to the loop with flanges. The main loop and its flanges are made of SUS316L type stainless steel. The hydrogen re-

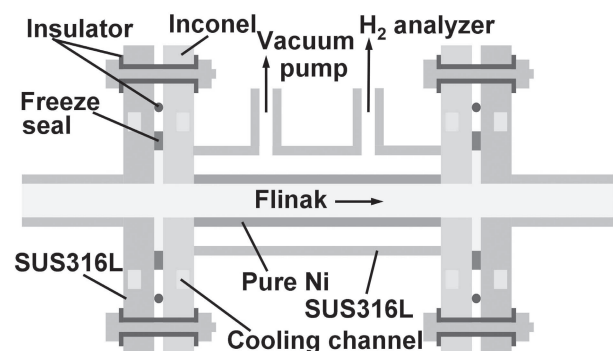


Fig. 1 Conceptual design for the hydrogen recovery unit for Orosh<sup>2</sup>i-1 loop.

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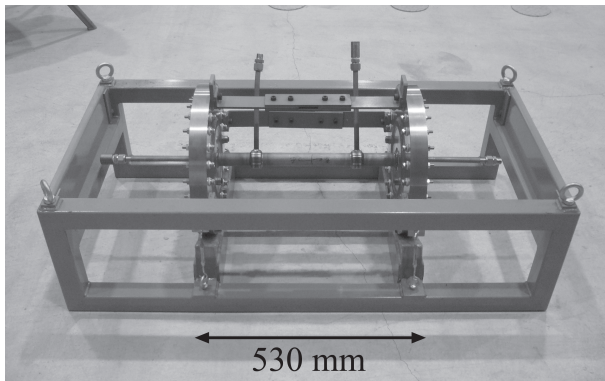


Fig. 2 The hydrogen recovery unit for Orosh<sup>2</sup>i-1 loop.

covery part is made of pure Ni tube with a grade Ni-200 and a purity of 99.6%. The outer diameter, wall thickness and length of the pure Ni tube are 12.7 mm, 0.8 mm, and 468 mm, respectively. The pure Ni tube was welded to Inconel 718 type Ni-base super alloy (52Ni-20Fe-19Cr-5Nb-3Mo-1Ti-0.5Al) flanges. Flinak flows inside the Ni tube. Electrochemical corrosion has been reported for JLF-1 ferritic steel contact with pure Ni in Flibe molten salt [7]. According to the electrochemical corrosion, Fe was dissolved from the steel, transferred to, and precipitated on Ni surface. The electrochemical corrosion is also expected for SUS316L steel, pure Ni, and Flinak system. It is considered that the Fe precipitates form a layer on the Ni tube surface and change the hydrogen permeation property. In order to avoid the electrochemical corrosion, the Ni-base alloy flanges are selected for contact to the Ni tube, and are electrically insulated from the SUS316L flanges. Insulator plastic spacer and rubber O-ring was located at the screws on the flanges and between the flanges, respectively. The flanges are water-cooled by the cooling channel in them. The cooling induces solidification of Flinak. The solidified Flinak acts as freeze seal maintaining insulation, and is expected to seal for flowing Flinak. A SUS316L tube is located outside the Ni tube. The SUS tube is connected to a vacuum pump and hydrogen analyzer; i.e., a Quadrupole mass spectrometer (QMS), to measure hydrogen permeated through the Ni tube wall from flowing Flinak. Figure 2 shows the hydrogen recovery unit fabricated. Both the SUS tube ends are plugged in the figure; however, they are to be connected to Orosh<sup>2</sup>i-1 loop in future.

### 3. Hydrogen Permeation Tests

In order to evaluate the hydrogen recovery unit, hydrogen permeability was measured at 530 K. The Ni tube in the unit was filled with He-3%H<sub>2</sub> gas mixture. Gas pressure was 1.6 atm and 3.0 atm, which are equivalent to  $4.9 \times 10^3$  Pa and  $9.1 \times 10^3$  Pa in partial pressure of hydrogen gas ( $P_x$ ), respectively. The vacuum in the gap between the Ni tube and the SUS tube was better than  $1 \times 10^{-5}$  Pa. The counts for H<sub>2</sub> by QMS were converted to permeation

Table 1 Hydrogen permeability for the Ni tube wall in the hydrogen recovery unit at 530 K.

Partial pressure, $P_x$ / Pa	Permeation rate, $n_x$ / mol s <sup>-1</sup>	Permeability, $q$ / mol m <sup>-1</sup> s <sup>-1</sup> Pa <sup>-0.5</sup>
$4.3 \times 10^3$	$1.2 \times 10^{-9}$	$8.2 \times 10^{-13}$
$9.1 \times 10^3$	$2.1 \times 10^{-9}$	$1.1 \times 10^{-12}$

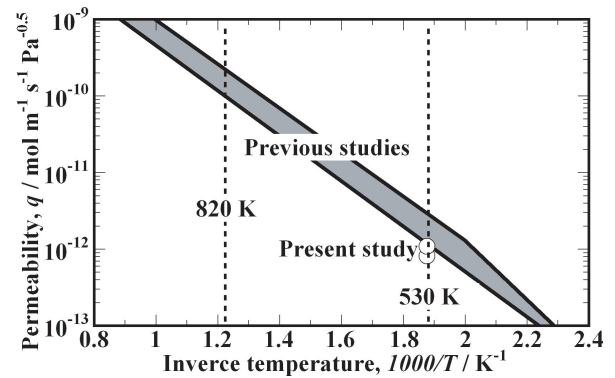


Fig. 3 Comparison of permeability of pure Ni for the hydrogen recovery unit and those in previous studies.

rate ( $n_x$ ). The permeation rate is converted to hydrogen permeability with the following,

$$q = \frac{n_x t_w}{S \sqrt{P_x}}, \quad (1)$$

where  $t_w$  is wall thickness (0.8 mm) and  $S$  is surface area ( $\pi \times 11.1$  mm  $\times$  468 mm) for the Ni tube. Table 1 lists the determined hydrogen permeability. Figure 3 plots the permeability obtained (circle symbol), compared with those in previous studies (gray zone) [8–15]. The present permeability is comparable to or slightly less than those in the previous studies. Possible reason for reduction in permeability is oxidation of the surface of the Ni tube during the welding with the Inconel flange in the air.

Unfortunately, 530 K is the maximum temperature for the current experiment system. The Ni tube is heated by a heater located outside the outer SUS316L tube. Vacuum insulating zone lies between the Ni and the SUS316L tubes; therefore, much heating and cooling power is required to keep both the Ni tube temperature at 820 K, and the rubber O-ring temperature lower than the melting point (450 K). Higher temperature test is planned after installation of the unit to Orosh<sup>2</sup>i-1 loop, where the Ni tube is heated by the flowing Flinak at 820 K. The degradation of the hydrogen permeability at 820 K would be similar or smaller than that at 530 K, if it is caused by the surface oxidation, because the oxidation layer is generally decomposed due to oxygen diffusion into Ni matrix at higher temperature under high vacuum condition. Although more data acquisition is required, significant degradation is not expected at 820 K.

### 4. Discussion for Hydrogen Recovery System

Outlet temperature of molten salt Flibe blanket in fusion reactor is 820 K [16]. Hydrogen permeability for pure Ni at 820 K has been already obtained as  $1 \times 10^{-10} \text{ mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$  as shown in Fig. 3. On the other hand, permeation data for the advanced hydrogen permeation materials (i.e., Pd-, V-, Nb-, and Ta-based alloys) have been limited below 670 K [3–6]. More investigation on permeability of the advanced materials is required especially at elevated temperatures, to discuss the application of them to hydrogen recovery system in fusion reactor.

Permeability can be roughly estimated from a product ( $q_e$ ) of diffusion coefficient ( $D$ ) and solubility ( $S_1$ ).

$$q_e = \frac{DS_1}{N_A}, \tag{2}$$

where  $N_A$  is Avogadro constant ( $6.02 \times 10^{23}$ ). Table 2 indicates the estimated permeability. This implies that potential permeability for Pd is expected as 100 times larger, and those for V, Nb, and Ta to be 1000 times larger than for Ni. The estimated permeability for Ni in Table 2 is similar to measured value in Fig. 3. Since  $10^{-8} \text{ mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$  or more has been reported as permeability even at 673 K for the advanced permeation materials [3–6], and generally increases with increasing temperature, the estimation in Table 2 seems to be reasonable.

Assuming that permeability of the hydrogen permeation materials in the hydrogen recovery unit is  $10^{-10}$  (Ni),  $10^{-8}$  (Pd), and  $10^{-7}$  (V, Nb and Ta)  $\text{mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$  at 820 K, the size for the hydrogen recovery system can be estimated. If molten salt is flowing through a tube, hydrogen permeation flux ( $Q$ ) is described as follows with the situation in Fig. 4,

$$Q = \frac{q(\sqrt{P} - \sqrt{P_v})}{t_w} dS = \frac{\pi D_t q(\sqrt{P} - \sqrt{P_v})}{t_w} dL, \tag{3}$$

where hydrogen gas layer with hydrogen pressure,  $P$ , is assumed at the interface between the flowing Flibe and the tube wall.  $P_v$  is a vacuum outside the tube,  $D_t$  is inner diameter of the tube, and  $L$  is the displacement from the tube inlet. The hydrogen pressure ( $P$ ) is obtained by Henry’s law with the hydrogen concentration (molar fraction) in Flibe ( $C$ ),

$$P = \frac{C}{h} = k C, \tag{4}$$

where,  $h$  is Henry’s constant for hydrogen in Flibe ( $2.13 \times 10^{-9} \text{ Pa}^{-1}$ ), and  $k$  is its inverse. Assuming Flibe is flowing with the flow velocity,  $v$ ,  $dC$  is the reduction in hydrogen concentration in Flibe due to the permeation flux,  $Q$ , during the time,  $dt$ , where Flibe is traveling from the displacement,  $L$ , to  $L + dL$ . Here, surface area for permeation flux ( $Q$ ) and volume for the concentration change ( $dC$ ) are  $dS$  and  $dV$ , respectively. The concentration change ( $dC$ ) is molar fraction of the lost hydrogen

Table 2 Diffusion coefficient ( $D$ ), solubility ( $S_1$ ) and their products ( $q_e$ ) at 820 K for pure metals for hydrogen permeation materials.

Element	Diffusion coefficient, $D / \text{m}^2 \text{ s}^{-1}$	Solubility, $S_1 / \text{m}^{-3} \text{ Pa}^{-0.5}$	Permeability, $q_e / \text{mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$
Ni	$3.2 \times 10^{-9}$	$2.1 \times 10^{22}$	$1.1 \times 10^{-10}$
Pd	$1.5 \times 10^{-8}$	$1.6 \times 10^{24}$	$4.0 \times 10^{-8}$
V	$1.7 \times 10^{-8}$	$1.0 \times 10^{25}$	$2.8 \times 10^{-7}$
Nb	$1.1 \times 10^{-8}$	$1.7 \times 10^{25}$	$3.1 \times 10^{-7}$
Ta	$8.3 \times 10^{-9}$	$3.0 \times 10^{25}$	$4.1 \times 10^{-7}$

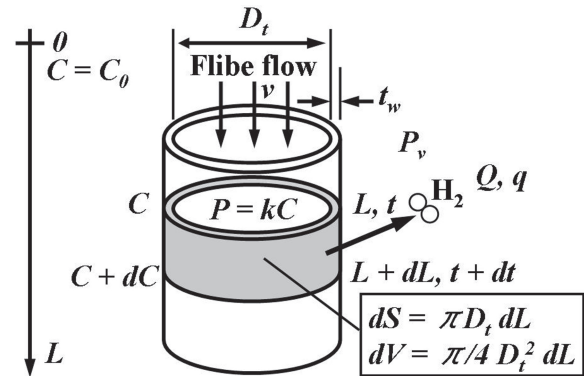


Fig. 4 Hydrogen permeation from Flibe through tube wall made of hydrogen permeation materials.

( $-Qdt$ ) against the Flibe amount in the volume ( $\rho dV/M$ ), where  $\rho$  is mass density of Flibe ( $2.0 \text{ g cm}^{-3}$ ), and  $M$  is molecular weight of Flibe ( $98.894 \text{ g mol}^{-1}$ ),

$$dC = -\frac{Q dt}{\frac{\rho dV}{M}} = -\frac{4 Q M dt}{\rho \pi D_t^2 dL} = -\frac{4 Q M}{\rho \pi D_t^2 v}. \tag{5}$$

The equations, (3), (4), and (5), are combined as,

$$\int \frac{dC}{\sqrt{k C} - \sqrt{P_v}} = -\int \frac{4 M q}{\rho D_t v t_w} dL. \tag{6}$$

The solution in the condition,  $C = C_0$  at  $L = 0$ , where  $C_0$  is the initial concentration at the tube inlet, is described as,

$$\frac{\sqrt{C_0} - \sqrt{C}}{\sqrt{k}} + \frac{\sqrt{P_v}}{k} \ln \frac{\sqrt{k C_0} - \sqrt{P_v}}{\sqrt{k C} - \sqrt{P_v}} = \frac{4 M q}{\rho D_t v t_w} L. \tag{7}$$

Figure 5 draws concentration-displacement curves for Flibe in the tube made of hydrogen permeation materials. Considering the possible reduction of the permeability in the present study indicated in Fig. 3, a curve for  $q$  equal to  $10^{-11} \text{ mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$  (1/10 of Ni) is also added.  $D_t$  and  $t_w$  were the same as the present hydrogen recovery unit; i.e., 11.1 mm and 0.8 mm, respectively.  $C_0$ ,  $v$ , and  $P_v$  were assumed as 1 mol ppm,  $1 \text{ m s}^{-1}$ , and  $7.6 \times 10^{-4} \text{ Pa}$

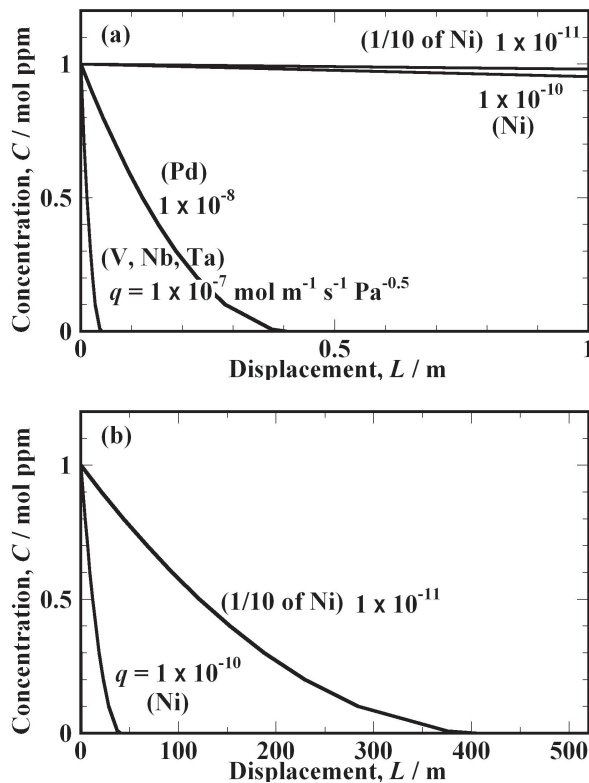


Fig. 5 Change in hydrogen concentration in Flibe flowing in a tube made of hydrogen permeation materials with various hydrogen permeability. Fig. (b) plots the concentration-displacement curves for  $q$  equal to  $1 \times 10^{-10}$  and  $1 \times 10^{-11} \text{ mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$  in Fig. (a) again with reduced horizontal axis.

( $1 \times 10^{-6}$  Torr), respectively, for the probable condition in fusion reactor. It is indicated that about 0.04, 0.4, and 40 m in tube length are required for the hydrogen permeation materials with the permeability of  $10^{-7}$  (V, Nb, and Ta),  $10^{-8}$  (Pd), and  $10^{-10}$  (Ni)  $\text{mol m}^{-1} \text{ s}^{-1} \text{ Pa}^{-0.5}$ , respectively. It is revealed that 10 times higher permeability results in 10 times shorter tube for hydrogen recovery. If the permeability is degraded during the fabrication process by 10 times (1/10 of Ni), tube length would be about 400 m. If 400 of 1 m-long tubes are located in 20 lines and 20 lines with an interval of 2 mm,  $0.3 \text{ m} \times 0.3 \text{ m} \times 1 \text{ m}$  is the size of the hydrogen recovery unit. The size is feasible for fabrication even in the case of 1/10 of Ni for hydrogen permeability. However, pressure drop for Flibe flow increases with increasing the tube length, and may limit the length of the hydrogen permeation tubes.

The above model assumed that the transfer of hydrogen in Flibe was much higher than that in hydrogen perme-

ation materials. This condition is equivalent to large flow stirring of Flibe. Analysis for thermodynamics of Flibe including hydrogen transfer is needed for estimation of performance of the hydrogen recovery unit. In addition, the present model assumed the hydrogen gas layer at the interface between Flibe and the tube wall controlled by Henry's law with Eq. (4). If hydrogen is transferred from Flibe to the hydrogen permeation materials in direct, the permeability would be changed probably due to change in solubility of hydrogen in the materials. Moreover, hydrogen permeability may change from Eq. (3) under high hydrogen concentration condition especially for hydrogen absorbing metal, such as V, Nb and Ta, if much hydrogen is accumulated in molten salt. Typical pressure-composition isotherm (PCT) curve and permeability diagram have been determined in the vacuum or gas atmosphere. Data accumulation for PCT curves and permeation diagrams in molten salt is necessary to investigate such direct hydrogen transfer between molten salt and the hydrogen permeation materials.

## 5. Conclusion

Hydrogen recovery unit for Orosh<sup>2</sup>i-1 loop was successfully fabricated with pure Ni tube as hydrogen permeation material, and showed comparable or little less hydrogen permeability to previous data for pure Ni. Advanced hydrogen permeation materials; e.g., Pd, V, Nb, and Ta, are also considered for hydrogen recovery system in hydrogen recovery systems in fusion reactor. 10 times higher hydrogen permeability potentially results in 10 times shorter length for hydrogen recovery tubes, thus much compact hydrogen recovery system can be expected.

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