Bi-Directional Hydrogen Isotopes Permeation through the First Wall of a Magnetic Fusion Power Reactor

Haishan ZHOU[1)]*, Yoshi HIROOKA[1,2)], Naoko ASHIKAWA[1,2)] and Takeo MUROGA[1,2)]

[1)]*Department of Fusion Science, The Graduate University for Advanced Studies, Toki 509-5292, Gifu, Japan*

[2)]*National Institute for Fusion Science, 322-6 Oroshi, Toki 509-5292, Gifu, Japan*

**Present address: Institute of Plasma Physics, CAS, P.O. Box 1126, Hefei, 230031, China*

(Received: 25 September 2014 / Accepted: 19 January 2015)

Plasma-driven permeation (PDP) and gas-driven permeation (GDP) through a first wall candidate material F82H have been investigated using a laboratory-scale steady state plasma facility. Experiments indicate that GDP may take place in the opposite direction of PDP, which then results in an unwanted increase in edge plasma density. A hydrogen flow from the plasma side to the gas side has also been identified. A one-dimensional diffusion analysis code, DIFFUSE, has been utilized to simulate the experiments. Multiple hydrogen isotopes (D/T) bi-directional permeation has been studied by DIFFUSE-code modelling as well. The two isotopic (D/T) flows appear to be independent of each other, driven by their own concentration gradients.

Keywords: plasma-driven permeation, gas-driven permeation, reduced activation ferritic steel, first wall, blanket, fusion power reactor

1. Introduction

The first wall of a fusion power reactor is defined as the plasma-facing surfaces of the blanket units, which are required to operate at elevated temperatures for an efficient heat exchange. For the blankets employing self-cooled breeder, the first wall is exposed to the edge plasma, containing energetic D^+ and T^+ ions on the one side and on the other side it is exposed to T_2 gas bred in blankets. Under these conditions, it is highly possible that these hydrogen isotopes would penetrate the first wall by a phenomenon called "bi-directional permeation": (1) deuterium as well as tritium would be transported into the blanket by plasma-driven permeation (PDP), which will probably necessitate isotope separation; and (2) tritium would flow in the counter direction to the edge plasma by gas-driven permeation (GDP), which will affect edge plasma density [1,2].

For the hydrogen isotopes permeation through the first wall, the fundamental materials science questions that need to be addressed are: (1) can plasma-driven and gas-driven permeation of hydrogen isotopes in the two counter directions actually take place under fusion reactor relevant conditions; and (2) will hydrogen isotopes in these two flows "recognize" each other? If they do, what would the resultant flows be like? To answer the first question, bi-directional hydrogen permeation experiments have been performed by exposing membrane samples to plasma and gas simultaneously. A one-dimensional diffusion code: DIFFUSE [3,4] has been utilized to simulate the experiments. The second question is related to the isotope effects on hydrogen permeation. For D-T fusion reactor studies, isotope effects must be taken into account because both D and T are the fuels. Isotope effects on hydrogen transport can be divided into two classes [5], i.e., intrinsic effects and synergistic effects. The former class is related to the differences in the transport properties of each of the individual isotopes. The latter class is due to the competition of the various isotopes for traps and the coupling of isotopes through the process of surface recombination. In the present work, the evaluation using DIFFUSE code has been extensively performed to deal with the multiple hydrogen isotopes (D, T) permeation cases as well.

2. Experimental

Details of the experimental setup in the steady state ECR plasma facility: VEHICLE-1 have already been presented in our previous papers [1,2]. For completeness, some of the important features of the bi-directional experiments are described in this section.

Shown in Fig. 1 is the schematic diagram of the bi-directional permeation setup in VEHICLE-1. F82H membranes are fixed in such a way that the upstream surface is exposed to hydrogen plasma, while the other side is exposed to hydrogen gas. At the plasma side, an orifice has been installed so that a quadrupole mass spectrometer (QMS) can measure the H_2 partial pressure in the VEHICLE-1 main chamber within the operational pressure. The plasma near the membrane surface is monitored by an optical spectrometer. For the PDP experiments, a bias of

haishanzhou@ipp.ac.cn

100 V or 50 V is applied and the electron density is of the order of 10^{16} /m^3. The electron temperature is raised up to ~10 eV for the improved sensitivity of H_α spectroscopy. Under such experimental conditions, the variation in surface recycling condition can be detected. At the gas side, the hydrogen gas pressure is measured by an absolute pressure gauge.

The F82H samples are prepared in the same dimensions as those commercially available conflat flanges with an outer diameter of 0.07 m, except that a circular area of ~0.035 m in diameter inside the knife-edge is machined down to thicknesses of 5×10^{-4} to 5×10^{-3} m to use as permeation membranes. Plasma-facing surfaces of the membranes are mechanically polished and then cleaned in an ultrasonic bath. During the experiments, the sample membranes can be heated up to >773 K by plasma bombardment and heat from a resistive heater.

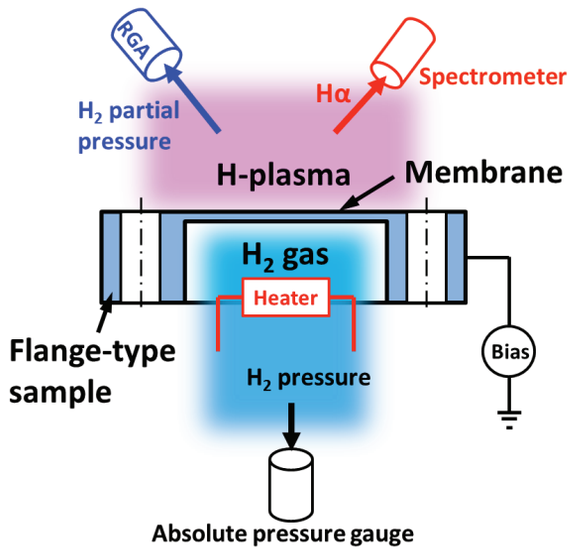

Fig.1 A schematic diagram of the bi-directional permeation setup in VEHICLE-1.

3. Bi-directional hydrogen (H) permeation demonstration and modelling

3.1. Hydrogen flow from the gas side to the plasma side (GDP flow)

For this experiment, a 6×10^{-4} m thick F82H membrane is used. The temperatures on the gas-facing and plasma-facing sides are measured to be ~853 and ~823 K, respectively. The ion bombarding energy is set at 50 V. Taking into account the ion species mix in the low temperature hydrogen plasma and the particle reflection at the plasma-facing surface, the net implantation flux is estimated to be ~8.5×10^{19} H /m^2/s.

Gas-driven permeation of hydrogen has been identified in the upstream hydrogen plasma, as shown in Fig. 2. The plasma is produced throughout the experiments and the downstream side is kept in high vacuum during the first ~100 s. From t = ~100 s, H_2 gas is introduced into the downstream to a pressure of 9.3×10^4 Pa to induce GDP (as shown in Fig. 1). Note that both the P_{H2} and H_α increase. Also seen here is the initial transient kick-up, which is exhibited by both P_{H2} and H_α, presumably due to ion-induced desorption or thermal desorption, although the detail is unclear at this point. At t = ~350 s, the H_2 gas is pumped out again and the P_{H2} and H_α decrease to the initial level. The steady state GDP hydrogen flow rate in this case has been evaluated to be about ~9.9×10^{19} H-atoms/m^2/s.

Figure 3 shows the hydrogen release flux from the plasma-facing surface predicted by the DIFFUSE code. For the DIFFUSE calculation, the input data are exactly the same as the experiment (e.g., membrane thickness, temperature, plasma flux, etc.), except that α-Fe is used as a surrogate of F82H because unfortunately F82H is not included in this database. The boundary conditions for the plasma side and the gas side are set to be recombination release and Sieverts' law, respectively. In the first 100 s, only plasma is produced. The hydrogen flux reaches ~8.5×10^{19} H-atoms/m^2/s (i.e., hydrogen recycling rate

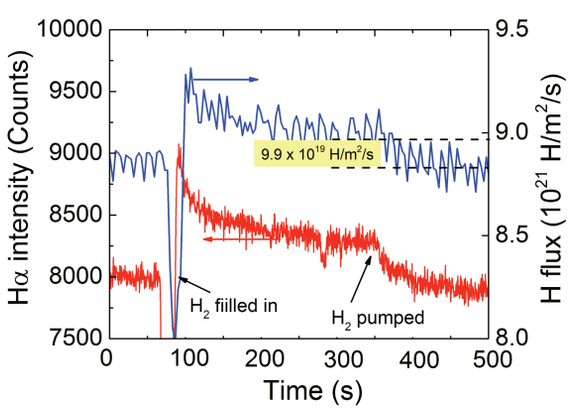

Fig.2 Hydrogen flux and H_α signals detected in the upstream hydrogen plasma in VEHICLE-1. Data rearranged from those published in [11].

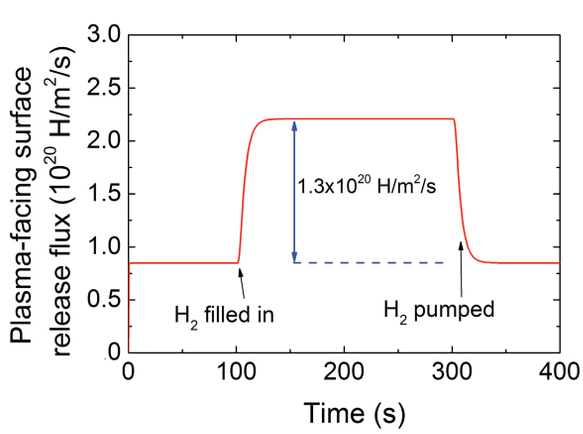

Fig.3 Hydrogen release flux from the plasma facing surface of the 6×10^{-4} m thick membrane predicted by the DIFFUSE code.

reaches ~100%) within one second. From 100 s to 300 s, hydrogen gas is introduced to the plasma downstream side. An extra steady-state release flux of 1.3×10^{20} H/m^2/s has been found, suggesting GDP takes place in the opposite direction of PDP. The hydrogen gas for driving GDP is pumped out from t = 300 s and the steady-state release flux is found to decrease to the initial value in the first 100 s. The estimated steady-state hydrogen GDP flux is relatively close to the experimental data.

3.2. Hydrogen flow from the plasma side to the gas side (PDP flow)

Hydrogen PDP flow from the plasma side to the gas side has been investigated as well. The results shown in the previous section indicate a net hydrogen flow from the gas side to the plasma side in the bi-directional permeation experiments. That means the hydrogen gas pressure at the PDP downstream will keep decreasing due to the loss of hydrogen. Then the PDP signal may be picked up from the decreasing rate of hydrogen pressure at the downstream side of the membrane. The total remaining hydrogen particle numbers in a closed volume at time t are given as $N_1(t)$ and $N_2(t)$ for the cases with and without PDP as:

$$N_0 - N_t - J_{\mathrm{GDP}}ts + J_{\mathrm{PDP}}ts = N_1(t) \quad (1) \text{ and}$$
$$N_0 - N_t - J_{\mathrm{GDP}}ts = N_2(t), \quad (2)$$

where N_0 is the initial particle number, N_t is the number of particles trapped in the membrane, s is the surface area, J_{GDP} and J_{PDP} are the steady state GDP and PDP flux, respectively. The PDP flux can be evaluated by:

$$J_{\mathrm{PDP}} = \frac{1}{s}\left(\frac{dN_1(t)}{dt} - \frac{dN_2(t)}{dt}\right). \quad (3)$$

Based on the above idea, bi-directional hydrogen permeation experiments have been performed with and without a bias of -100 V onto the sample membrane. Our previous experiments indicate that hydrogen PDP flux is much higher when the bias is applied [6]. Here the PDP flow is assumed to be negligible when the bias is off, i.e., only GDP. If a PDP flow into the gas side actually exists in the bi-directional permeation process, the decreasing rate of hydrogen gas pressure at the downstream side should be smaller than that of the GDP-only case.

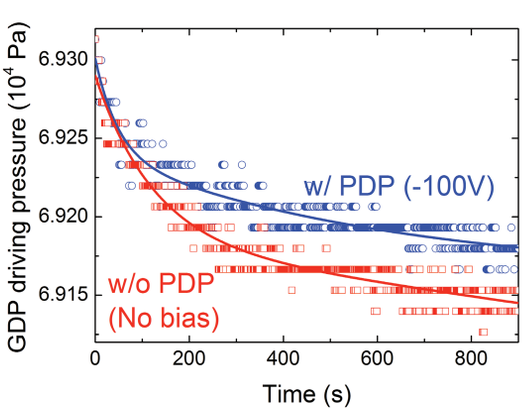

Fig.4 Variation of hydrogen gas pressures measured by an absolute pressure gauge at the PDP downstream side.

Hydrogen gas is introduced into a closed volume (PDP downstream) with an initial hydrogen gas pressure of ~6.93×10^4 Pa. Figure 4 shows the time evolution of hydrogen gas pressures for the two cases (i.e., w/ and w/o PDP). It can be seen that after the transient phases, both of the hydrogen pressures decrease, indicating hydrogen outflows to the plasma side. The difference in the two curves reflects the effect of PDP.

Using Eq. (3) and the steady state portion of the curves shown in Fig.4, the PDP flux is estimated to be ~2.3×10^{18} H/m^2/s, which is about 30% higher than theoretical prediction for steady state PDP flux in the recombination-diffusion limited regime [7]:

$$J_{\mathrm{PDP}} = \frac{D}{L}\sqrt{\frac{J_0}{K_r}} \quad (4),$$

where J_0 is the net implantation flux; L is the membrane thickness; D and K_r are the diffusion coefficient and surface recombination coefficient for F82H [8]:

$$D = 7.5\times10^{-4}\exp\left(\frac{-0.14\ \mathrm{eV}}{kT}\right) \quad (5)$$
$$K_r = 4.8\times10^{-21}\exp\left(\frac{0.48\ \mathrm{eV}}{kT}\right). \quad (6)$$

4. Modelling on reactor operation with hydrogen isotopes (D,T) bi-directional permeation through the first wall

4.1. Re-evaluation of tritium pressure in a FLiBe blanket

The T_2 equilibrium pressure in FLiBe is available [9]. Assuming a tritium concentration of 0.1 ppm, the T_2 pressure is estimated to be ~10^4 Pa at 800 K. However, fusion reactor studies indicate that the T_2 gas pressure in a blanket is related to a number of factors, e.g., fusion power, tritium breeding ratio, tritium recovery rate, structure material, temperature, flow rate of the breeder, etc.

Evaluation of the T_2 gas pressure in the tritium loop was reported in another paper [10], assuming no T leakage from the first wall. Under these conditions, the pressure was found to be 10^3-10^4 Pa, depending on the integration method of the tritium recovery system. Preliminary tritium analysis for the blanket employing FLiBe was performed as well [11]. In the presence of tritium permeation barrier, the T_2 pressure varied from 5×10^3 to 4.5×10^4 Pa as a decrease of tritium recovery efficiency. To analyze the

hydrogen isotopes permeation through a bare first wall made by a reduced activation ferritic steel alloy, re-evaluation of the tritium pressure has been performed in the present work.

The overall tritium balance in the loop is given by:

$$\frac{\partial M_T}{\partial t} = J_1 + J_2 - \sum J_{3-i} - J_4 - J_5, \qquad (7)$$

where M_T is the tritium inventory in the loop; J_1 is the tritium production rate; J_2 is the tritium flow towards the blanket by plasma-driven permeation; J_3 is tritium leakage due to gas-driven permeation, including tritium leakage from the first wall ($J_{3\text{-wall}}$) and the pipes ($J_{3\text{-pipe}}$); J_4 is the extracted tritium by the tritium recovery system; J_5 is the tritium leakage in the heat exchanger. Tritium decay is neglected in the present analysis.

For the blanket of a 3 GW fusion reactor with a tritium breeding ratio (TBR) of 1.3 [12], the tritium breeding rate J_1 is estimated to be 1.38×10^{21} T-atoms/s (6.89×10^{-3} g/s T). The hydrogenic ion implantation flux is assumed to be 1×10^{20} atoms/m^2/s (50% T and 50% D), which will lead to a steady state PDP flux of 1×10^{17} atoms/m^2/s , i.e., J_2 = 2.5×10^{-4} g/s T for a plasma facing surface area of 1000 m^2. For calculating the tritium leakage from the first wall by GDP, the total surface area of the blankets modules is 3000 m^2 for a helical type reactor [12] and the wall thickness L is 5×10^{-3} m [13]. The other input data for the calculation are as follows: the FLiBe flow rate is 2.2×10^6 m^3/s; the tritium solubility is 5.3×10^{-13} wt fraction/Pa [14]; the tritium recovery efficiency is 0.99; the surface area and thickness data for the coolant pipes and heat exchanger are the same as those used in literature [11].

Solving Eq. (7) for a steady state condition, the tritium pressure has been found to be ~1.1×10^3 Pa, which is only ~10% of the estimated tritium equilibrium pressure in FLiBe at a T concentration of 0.1 ppm at 800 K [9].

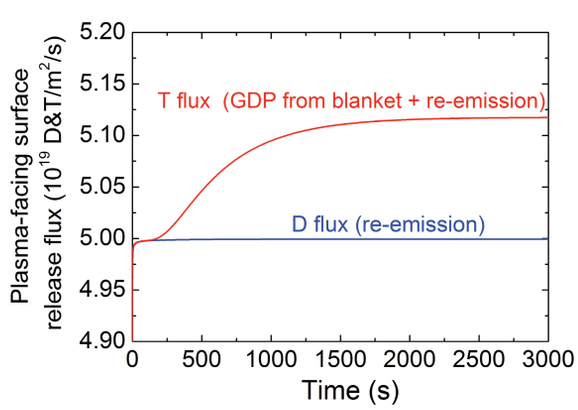

Fig.6 D and T release fluxes at the plasma-facing surface calculated by the DIFFUSE-code for bi-directional PDP-D/T and GDP-T_2. The total T flux is composed of the re-emission flux of implanted T particle and the GDP-T from the blanket side.

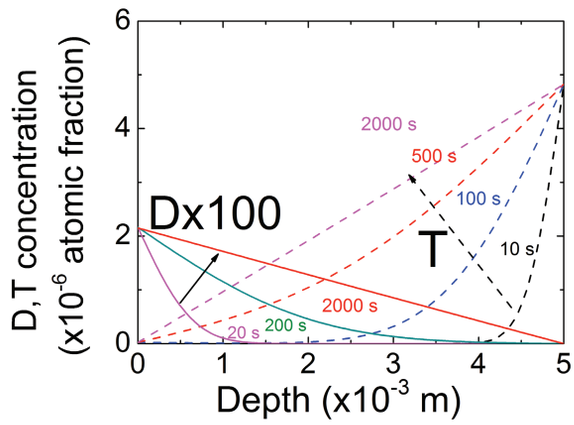

Fig.5 Time evolution of D/T-concentration profiles calculated by the DIFFUSE-code for bi-directional PDP-D/T and GDP-T_2 from the upstream and downstream surfaces, respectively. The D concentration is enlarged by 100 times.

4.2. Re-evaluation of hydrogen isotopes permeation through the first wall of FLiBe blankets

Using the tritium partial pressure data shown in section 4.1, hydrogen isotopes permeation through the first wall of FLiBe blankets has been re-evaluated by DIFFUSE-code as well.

The input data for DIFFUSE calculation are as follows: the first wall thicknesses are assumed to be 5×10^{-3} m and the implanted hydrogen fluxes (D/T) at the plasma-facing side are assumed to be 1×10^{20} atoms/m^2/s (5×10^{19} D/m^2/s and 5×10^{19} T/m^2/s) with a bombarding energy of 100 eV. At the blanket side, the tritium gas pressure is 1.1×10^3 Pa. The first wall temperature is assumed to be the average value of the inlet (773 K) and outlet (873 K) coolant temperatures. A trap density of 1% atomic fraction and a tapping energy of 0.62 eV are assumed.

Figure 5 shows the time evolution of the tritium and deuterium concentration profiles calculated by the DIFFUSE-code. Note that the T-concentration profile reflects two contributions: one by PDP from the upstream surface and the other by GDP from the downstream surface, whereas the D-concentration profile is only due to PDP. Most interestingly, the two isotopic (D/T) flows appear to be independent of each other, driven by their own concentration gradients. Here, note that the D concentration shown in Fig. 5 is enlarged by 100 times. The actual tritium concentration at the plasma-facing surface (depth ≈ 0) is larger than that of deuterium.

Figure 6 shows the D and T release fluxes at the plasma-facing surface calculated by the DIFFUSE-code.

The difference in T and D fluxes are mainly due to the T-GDP from the blanket side. The T-GDP flux is about 1.2×10^{18} T/m^2/s, which is 1.2% of the net implantation flux from plasma.

Taking hydrogen isotopes permeation into account, the first wall recycling rate R is defined as:

$$R = \frac{\Gamma_{reflection} + \Gamma_{re-emission} + \Gamma_{GDP}}{\Gamma_{plasma}}, \qquad (8)$$

where Γ_{plasma} is the total incident ion flux from edge plasma; $\Gamma_{\text{reflection}}$ is the particle flux by surface reflection; $\Gamma_{\text{re-emission}}$ is the particle flux by remission and Γ_{GDP} is the steady state plasma- gas-driven permeation flux. Assuming a particle reflection coefficient of 0.5, the total incident flux would be 2×10^{20} D&T/m^2/s and the first wall recycling rate has been estimated to be R = 1.006.

5. Conclusion

A laboratory-scale plasma device: VEHICLE-1 has been installed with an experimental setup for bi-directional hydrogen permeation by plasma-driven permeation (PDP) and gas-driven permeation (GDP) under conditions to be seen in the first wall environment.

Bi-directional hydrogen permeation through F82H has actually been demonstrated in a laboratory-scale steady state plasma facility. Both PDP flow to the gas side and GDP flow to the plasma side have been clearly identified.

Re-evaluation of the tritium flows in a FLiBe loop has been performed, taking into account tritium leakage from the first wall. The tritium equilibrium pressure has been estimated to be ~1.1×10^3 Pa. DIFFUSE-code modelling predictions indicate that the two isotopic (D/T) flows appear to be independent of each other, driven by their own concentration gradients. Under the bi-directional hydrogen permeation conditions, the first wall recycling rate has been estimated to be $R > 1$ for the blanket employing FLiBe as a coolant/breeder.

Unfortunately, at present deuterium cannot be used in our laboratory because of local regulation. Further validation on the bi-directional phenomena using multiple hydrogen isotopes is anticipated in the future.

Acknowledgement

The author would like to thank Mr. J. Yagyu, Dr. M. Sato, Dr. T. Nakano and Dr. A. Sakasai of JAEA, Naka for their material preparation.

References

[1] Y. Hirooka and H. Zhou, Fusion Sci. Technol. **66**, 63 (2014).

[2] H. Zhou, Y. Hirooka, N. Ashikawa, T. Muroga and A. Sagara, Plasma Fusion Res. **8**, 2402065 (2013).

[3] M. Baskes, *DIFFUSE83*, Sandia Report SAND83-8231.

[4] Y. Hirooka, H. Zhou, N. Ashikawa, T. Muroga and A. Sagara, Fusion Sci. Technol. **64**, 345 (2013).

[5] D. K. Brice, J. Nucl. Mater. **122&123**, 1531 (1984).

[6] H. Zhou, Y. Hirooka, N. Ashikawa and T. Muroga, *Hydrogen gas and plasma driven permeation in counter directions through the first wall of a DEMO reactor*, Paper presented at the 28th JSPF Annual Meeting, Kanazawa, Japan, Nov. 22nd-25th, 2011.

[7] B. Doyle, J. Nucl. Mater. **111&112**, 628 (1982).

[8] H. Zhou, Y. Hirooka, N. Ashikawa, T. Muroga and A. Sagara, J. Nucl. Mater. **455**,470 (2014).

[9] S. Fukada, Y. Edao, S. Yamaguti and T. Norimatsu, Fusion Eng. Des. **83**, 747 (2008).

[10] S. Fukada, R. Anderl, A. Sagara and M. Nishikawa, Fusion Sci. Technol. **48**, 666 (2005).

[11] Y. Song, A. Sagara, T. Muroga, Q. Huang, M. Ni and Y. Wu, Plasma Fusion Res. **7**, 245016 (2013).

[12] A. Sagara, T. Goto, J. Miyazawa, N. Yanagi, T. Tanaka, H. Tamura, R. Sakamoto, M. Tanaka, K. Tsumori, O. Mitarai, S. Imagawa, T. Muroga and the FFHR design group, Fusion Eng. Des. **87**, 594 (2012).

[13] A. Sagara, H. Yamanishi, T. Uda, O. Motojima, T. Kunugi, Y. Matsumoto, Y. Wu, H. Matsui, S. Takahasi, T. Yamamoto, S. Toda, O. Mitarai, S. Satake, T. Terai and S. Fukada, Fusion Technol. **39**, 753 (2001).

[14] S. Fukada, *A design for recovery of tritium from FLiBe loop in FFHR-2*, Japan-US Workshop on Fusion Power Plants and Fusion Power Plants and Related Advanced Technologies with Participation of EU, Kyoto, Japan, Feb. 5th, 2007.