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**TEST OF THE PALLADIUM DIFFUSER IN THE JAERI  
FUEL CLEANUP SYSTEM IN THE TRITIUM SYSTEMS  
TEST ASSEMBLY**

March 1993

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Test of the Palladium Diffuser in the JAERI Fuel Cleanup System  
in the Tritium Systems Test Assembly

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The JAERI Fuel Cleanup System (JFCU) is a major subsystem of the TSTA simulated fusion fuel loop. The palladium diffuser, that accepts simulated plasma exhaust and purifies the hydrogen isotopes mixture for the feed to the Isotope Separation System, was tested with deuterium to investigate the characteristics of the components. Permeation flow rate is a linear function of the difference of the square root of the pressure across the palladium alloy membrane. However at the low pressure region, an impediment on the permeation was observed. It was suspected to be caused by the impurity adsorbed on the surface of the permeated side of the membrane and was reduced by oxidation treatment.

Keywords: Nuclear Fusion, Tritium, Deuterium, TSTA, Fusion Fuel Cycle, palladium, Permeation, Diffusion, Isotope Effect, Oxidation

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## TSTA用原研製燃料精製システムのパラジウム拡散器試験

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(1993年3月8日受理)

原研は日米協力協定AnnexIVに基づいて米国ロスアラモス国立研究所のトリチウムシステム試験施設(TSTA)において核融合炉燃料ループの模擬試験を共同で行っており、その一環として原研製の燃料精製システムを設計、制作してTSTAに設置、結合した。この装置に於て水素同位体を精製するパラジウム拡散器の基本的な特性である透過性能を測定した。透過流量は概ね $1/2$ 乗則に従うが、透過側圧力の低いときにはある一定の圧力差までは透過が起こらない現象が見いだされた。これはパラジウム合金表面の不純物に起因するものと思われ、酸化処理によりその悪影響は減少した。

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## I. Introduction

The JAERI Fuel Cleanup System (JFCU) is a complete plasma exhaust processing subsystem designed and fabricated by JAERI for the test in the TSTA fuel loop. The apparatus of the JFCU was installed in early 1990 in the TSTA and a number of tests without tritium were performed since then to verify the function of the components. The palladium diffuser is a front end component of the process that accepts simulated plasma exhaust. Palladium Diffuser separates hydrogen -impurity mixture into a pure hydrogen product stream and a bleed stream where impermeable impurities are concentrated. The permeated side of the palladium membrane is evacuated by a Scroll pump to drive permeation and compress the product to desired pressure for supply to the ISS. Low suction pressure is desired to reduce hydrogen concentration in the diffuser exhaust "bleed", in which partial pressure of hydrogen is dominated by this pressure.

In order to verify the basic performance of the component, the permeability of hydrogen isotopes were measured with the palladium diffuser-scroll pump loop. This report describes the outline of the procedure and result of this test. The results is expected to provide some information on the characteristics of the component in a practical use and help the understanding on the behavior.

## II. Test Plan TTA-TP-118-08

PLAN FOR THE JAERI FUEL CLEANUP SYSTEM  
PALLADIUM DIFFUSER TEST

## 1. Purpose

This test plan describes the outline of the cold testing of the palladium diffuser loop of the JFCU (JAERI Fuel Cleanup System). The palladium diffuser produces pure hydrogen isotope stream ( $Q_2$ ) for the ISS. Major purpose of this test is to evaluate the permeability of the diffuser driven by the scroll pump and the additional metal bellows pump with  $H_2$  and  $D_2$ . No tritium is used. Throughput of hydrogen will be obtained as functions of the square root of the pressures at the inlet and outlet of the diffuser. Another major purpose is training and familiarization in JFCU operation.

## 2. Configuration

The test will be conducted with a loop consists of PD, SCROL, RT1, RT2 and the metal bellows pump to be installed in the loop.  $H_2$  and  $D_2$  will be supplied from an external source. The vacuum manifold of the JFCU will be used for vacuum service. An external vacuum pump will be used. The simplified test flow is in Fig.1. A detailed configuration is highlighted in the attached drawing.

## 3. Subsystem required

No TSTA subsystems are required to conduct the test except for MDAC and UTIL. High pressure nitrogen, chilled water, ventilation and electric power both from regular and UPS source are needed. Normal operations and maintenance of the TSTA subsystems during this testing may be conducted. MDAC will monitor and log the JFCU data.

## 4. Personnel

S. Konishi, M. Inoue, T. Watanabe, J. W. Barnes and W. Harbin will conduct the test. TSTA operators will be involved in the operation.

## 5. Time

The test will be conducted in the week of Nov. 5 and 12, as a part of the remainder of the Scroll pump tests. The test will take approximately two weeks.



## 6. Possible hazard

No tritium will be used in the test. Potential hazards are related to tens of liters of hydrogen gas. All the major hazards are monitored and alarmed and/or interlocked.

## 7. Outline

The test procedure is almost the same as in the test of the scroll pump except the flow path involves the PD while it was by-passed in the pump test. Therefore, refer to detailed procedure TTA-TP-118-02.

### 7.1 Preparation

Install the metal bellows pump as in the design change that has already been submitted for QA review. The test may be performed with the pump at the current location if necessary.

Turn on the Scroll pump oil circulation pump. Supply cooling water.

Evacuate the test loop

Turn on the heater of the PD.

### 7.2 Test

Fill the part of the loop with H<sub>2</sub> to ca. 300 torr as shown in the attached figure.

Open all the valves in the loop.

Run the scroll pump, and then metal bellows pump.

Measure following parameters when stabilized.

FR-VACTPU, FR-PDIN, FR-ISSIN, PRA-RT1, PRA-SCROLIN, PRCA-RT2,, PRCA-ISSIN.

Slightly close HV523 or 524 to add differential pressure across the PD.

Measure the parameters, and close the valve, and repeat until the valve is completely closed.

Add H<sub>2</sub> and repeat above procedure until the pressure in RT2 reaches 850 torr.

Change PD temperature to 450°C stepwise and repeat the measurement.

Replace H<sub>2</sub> with D<sub>2</sub> and repeat the test. Deuterium will be accounted.

### 7.3 Shut down

Turn off the pumps and open the bypass valve.

Recover D<sub>2</sub> with the ZCB1.

Evacuate the loop.

Turn off the heater of the PD.

## 8. Data

Permeation flow rate through the diffuser is measured as a function of differential pressure across the membrane and the temperature. Report will be written by S. Konishi. All the data and operation log will be recorded in the JFCU computers well as at MDAC archive.

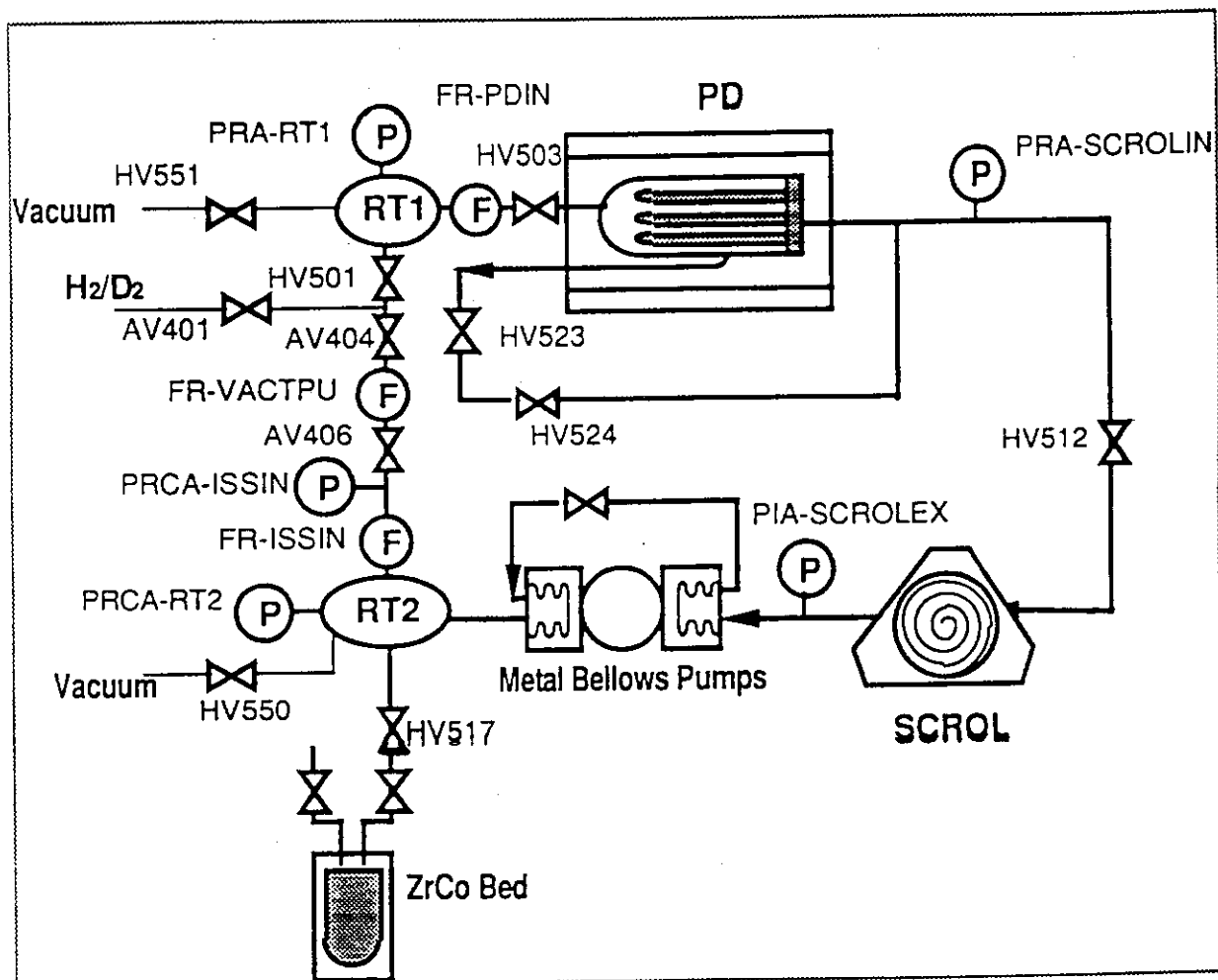
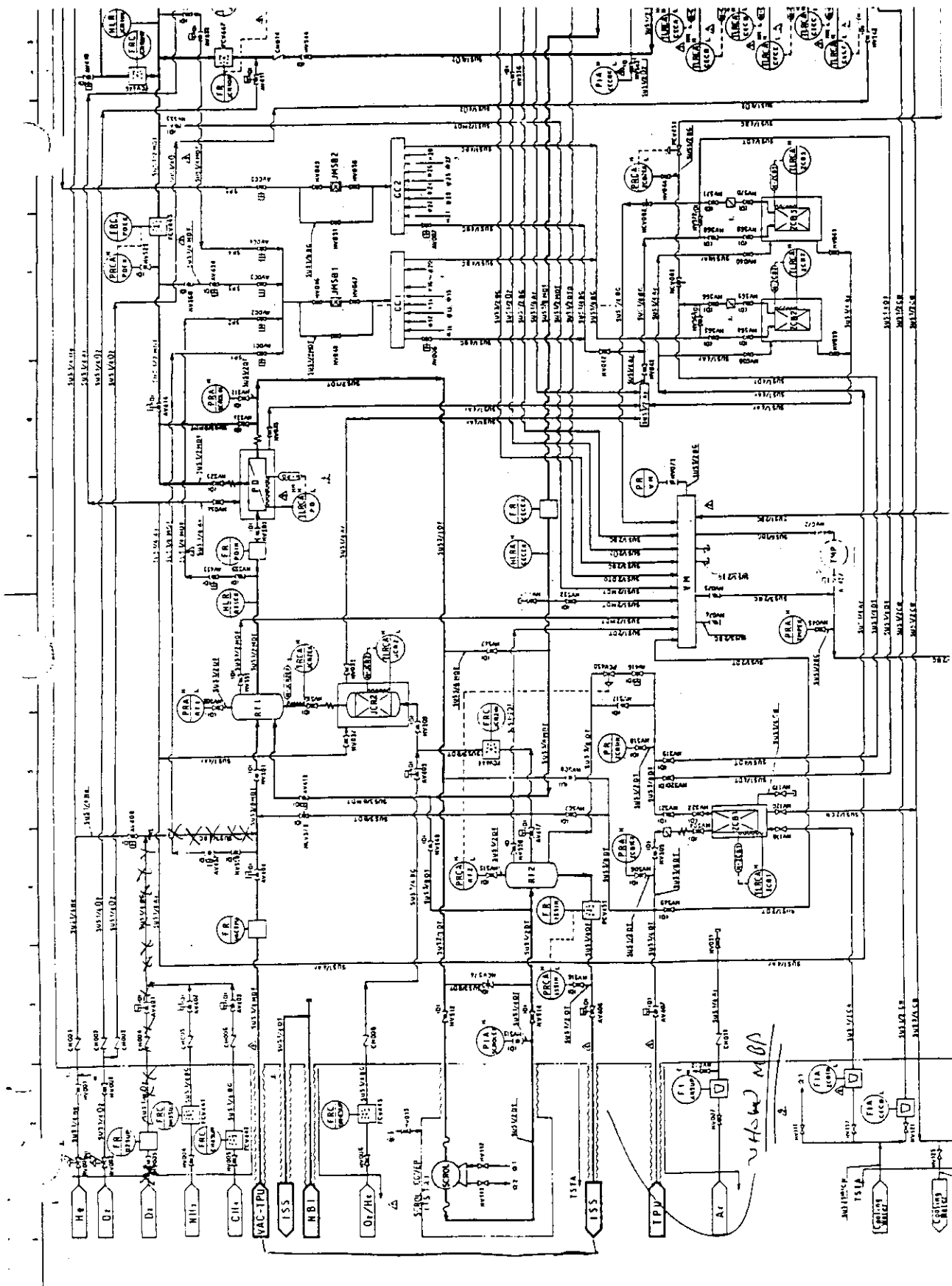


Fig. 1 Simplified test flow in the JFCU.



### III. Test Results

#### JFCU PD TEST REPORT

T. Watanabe

##### 1. Summary

THIS REPORT DESCRIBES THE RESULTS OF THE JFCU PD (PALLADIUM DIFFUSER) TEST.

THIS TEST HAS BEEN CONTINUED FOR FIVE WEEKS (NOV. 14 - DEC. 14, 1990). (THIS TERM OVERLAPPED WITH THAT OF INSTALLATION OF NEW CEC (CERAMIC ELECTROLYSIS CELL). IN THE CASE OF THE INTERVENTION, WE YIELDED THE TIME AND PLACE. SO THE TEST'S TERM WERE ABOUT 2 OR 3 WEEKS SUBSTANTIALLY.)

THE DIFFUSER WAS TESTED AT 573K AND 723K WITH H<sub>2</sub> AND D<sub>2</sub>. OBSERVED PERMEABILITY SATISFIES THE DESIGNED THROUGHPUT. A NORMETEX SCROLL PUMP AND A METAL BELLOWS PUMP WERE USED FOR CIRCULATION.

RELATIONSHIP BETWEEN THE PERMEATION FLOW RATE THROUGH PD AND THE DIFFERENCE OF SQUARE ROOT OF PRESSURES AT THE INLET AND OUTLET OF THE PD WAS OBTAINED. THE DEPENDENCE OF THE OBTAINED LINEAR FUNCTIONS ON THE PRESSURE AT THE PRODUCT SIDE WERE FOUND.

## For Your Information

##### 2. Purpose

THE MAJOR PURPOSE OF THIS TEST IS TO EVALUATE THE PERMEABILITY OF THE DIFFUSER DRIVEN BY THE SCROLL PUMP AND THE ADDITIONAL METAL BELLOWS PUMP WITH H<sub>2</sub> AND D<sub>2</sub>. ANOTHER PURPOSE IS TRAINING AND FAMILIARIZATION IN JFCU OPERATION.

##### 3. Test outline

THE FLOW PATH WE USED IS SHOWN IN FIG. 1.

THE TEST LOOP WAS EVACUATED BY OPENING HV550 AND HV551 TO THE ALCATEL PUMP. WE SUPPLIED H<sub>2</sub> (OR D<sub>2</sub>) TO THE TEST LOOP BY OPENING AV401. AFTER FILLING OF GAS, WE STARTED NEW METAL BELLOWS PUMP AND SCROLL PUMP. HCV574 WAS USED FOR THE CONTROL OF PRESSURE AT THE OUTLET OF PD.

THE FOLLOWING PARAMETERS WERE MAINLY MEASURED.

- PRA-RT1 ( PRESSURE AT THE INLET OF PD )
- PRA-SCROLIN ( PRESSURE AT THE OUTLET OF PD )
- FR-PDIN ( FLOW RATE AT THE PD )
- TLRCA-PD ( TEMPERATURE OF THE PD )

THE TEST'S PROCESS IS FOLLOWING.

- (1) NOV. 13, 14 PREPARATION
- BEFORE STARTING TEST, WE PREPARED AND CHECKED THE FOLLOWINGS.
- \* INSTALLATION OF THE NEW METAL BELLOWS PUMP AT DOWNSTREAM OF SCROLL PUMP
  - \* TURNING ON THE SCROLL PUMP OIL CIRCULATION PUMP
  - \* SUPPLY OF COOLING WATER FOR SCROLL PUMP
  - \* EVACUATION OF THE LOOP AND ZERO ADJUSTMENT OF THE PRESSURE GAGES AND THE FLOW METERS
  - \* TURNING ON THE HEATER OF THE PD

- #### 4. Test result

THE PERMEATION FLOW RATE FR-PDIN IS PLOTTED AGAINST THE DIFFERENTIAL SQUARE ROOT OF PRESSURE ACROSS THE MEMBRANE, AS A FUNCTION OF THE PRESSURE AT THE PERMEATED SIDE, PRA-SCROLIN.

THE COMPARISON BETWEEN FIG.2-1 AND FIG.2-3 ( Or FIG.2-2 AND FIG.2-4 ) SHOWS THAT THE PERMEABILITY IN H<sub>2</sub> IS APPROXIMATELY 1.5 TIMES AS HIGH AS THAT IN O<sub>2</sub> UNDER THE SAME TEMPERATURE.

THE DIFFERENCE OF SQUARE ROOT OF THE PRESSURE AT THE FEED SIDE

AND PERMEATED SIDE OF MEMBRANE WHEN THE PERMEATION FLOW RATE IS ZERO ( $DPO$ ) ARE PLOTTED AGAINST THE PRESSURE AT THE PERMEATION SIDE IN FIG.3-1 AND FIG.3-2 . (THE AXIS OF ORDINATE IS LOGARITHMIC SCALE.) THESE ARE ESTIMATED FROM THE LINER FUNCTION IN FIG.2-1 - FIG.2-5. THESE GRAPHS SHOW THAT THERE IS THE LINER FUNCTION BETWEEN TWO FACTORS. SO IT IS POSSIBLE TO EXPECT  $DPO$  (SEE ABOVE) AT ANY PRESSURE AT THE PERMEATED SIDE. AND FURTHER, IT IS POSSIBLE TO EXPECT THE LINER FUNCTION OF OTHER PRESSURE AT THE PERMEATION SIDE, IF IT IS SUPPOSED THAT THESE LINES OF FUNCTION ARE PARALLEL.





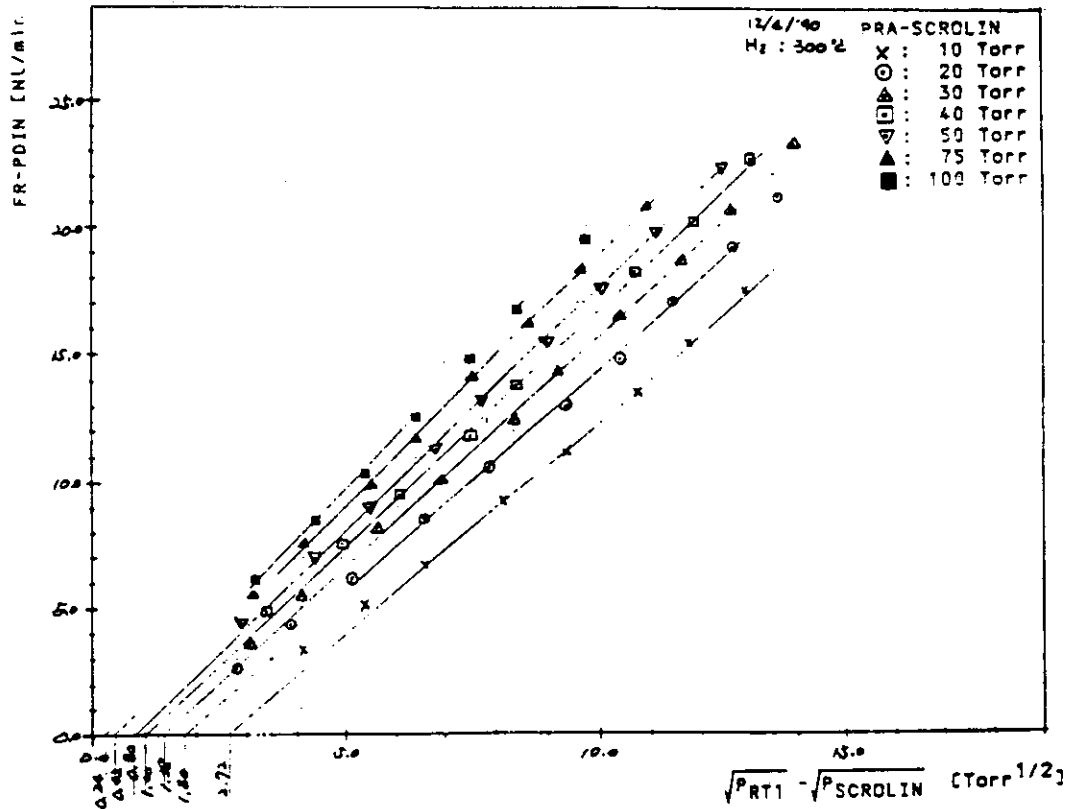


Fig. 2-1 Relationship between the permeation flow rate and the difference of square root of the pressure at the feed side and permeated side of membrane (H<sub>2</sub>, 573K)

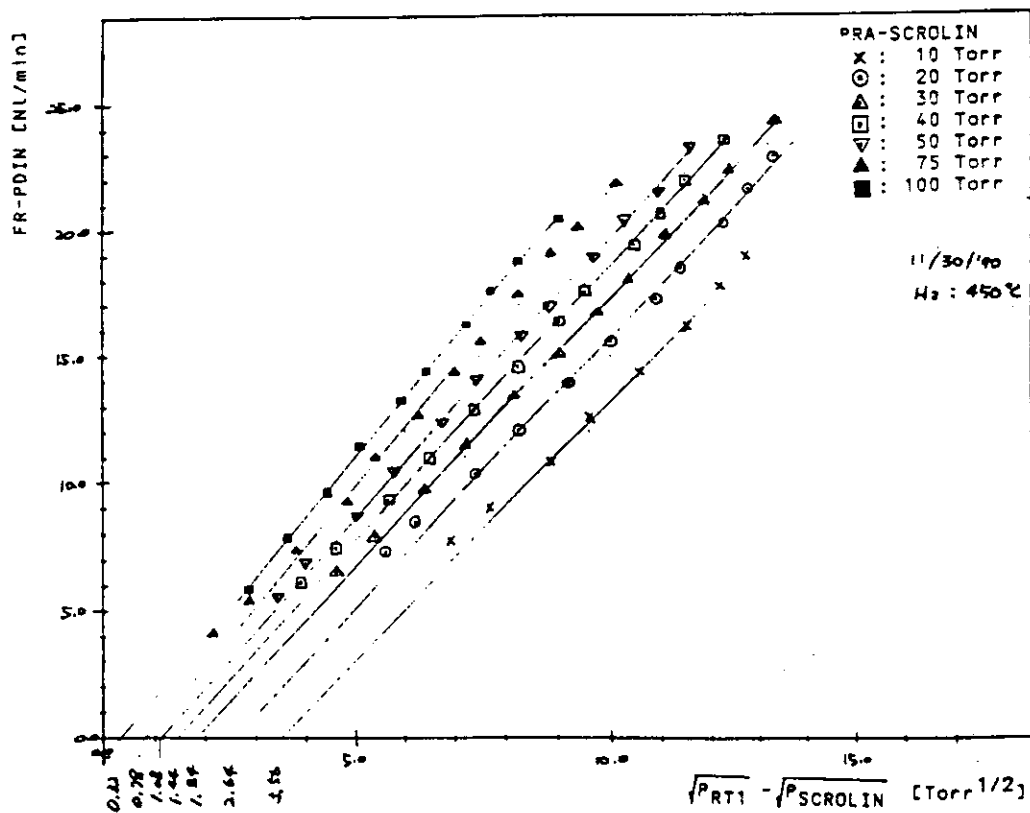


Fig. 2-2 H<sub>2</sub>, 723K

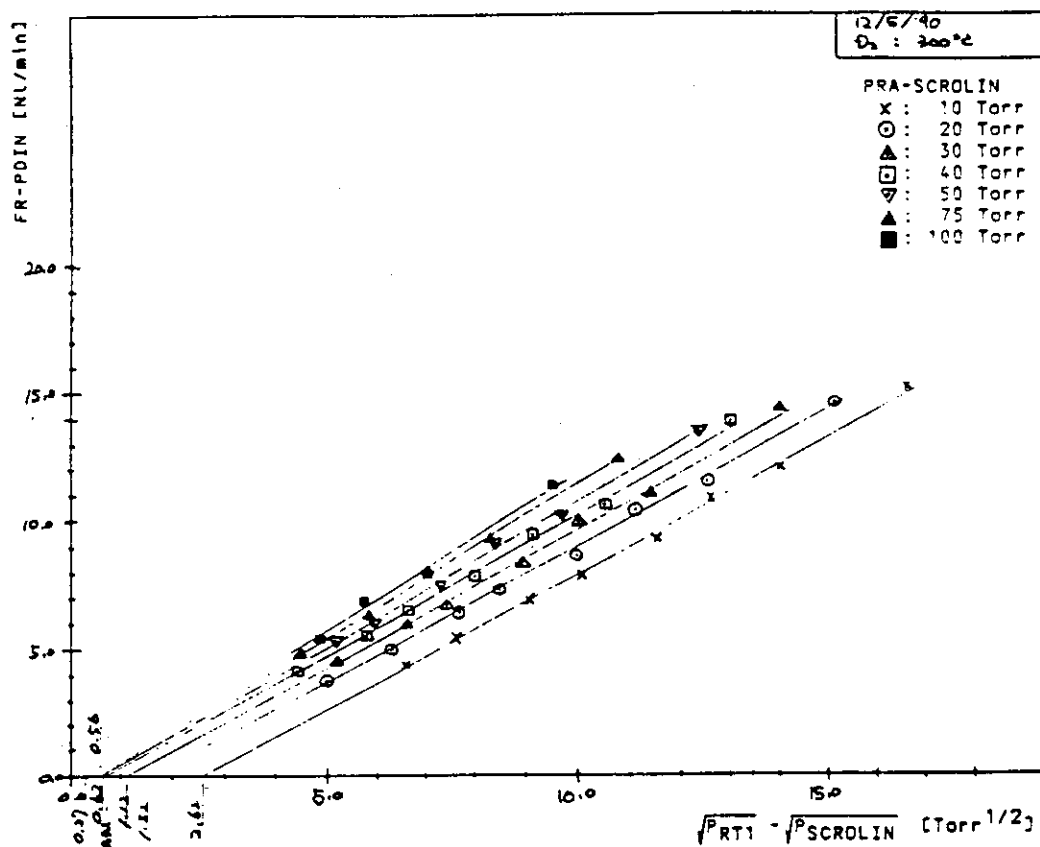


Fig. 2-3 D2,573K

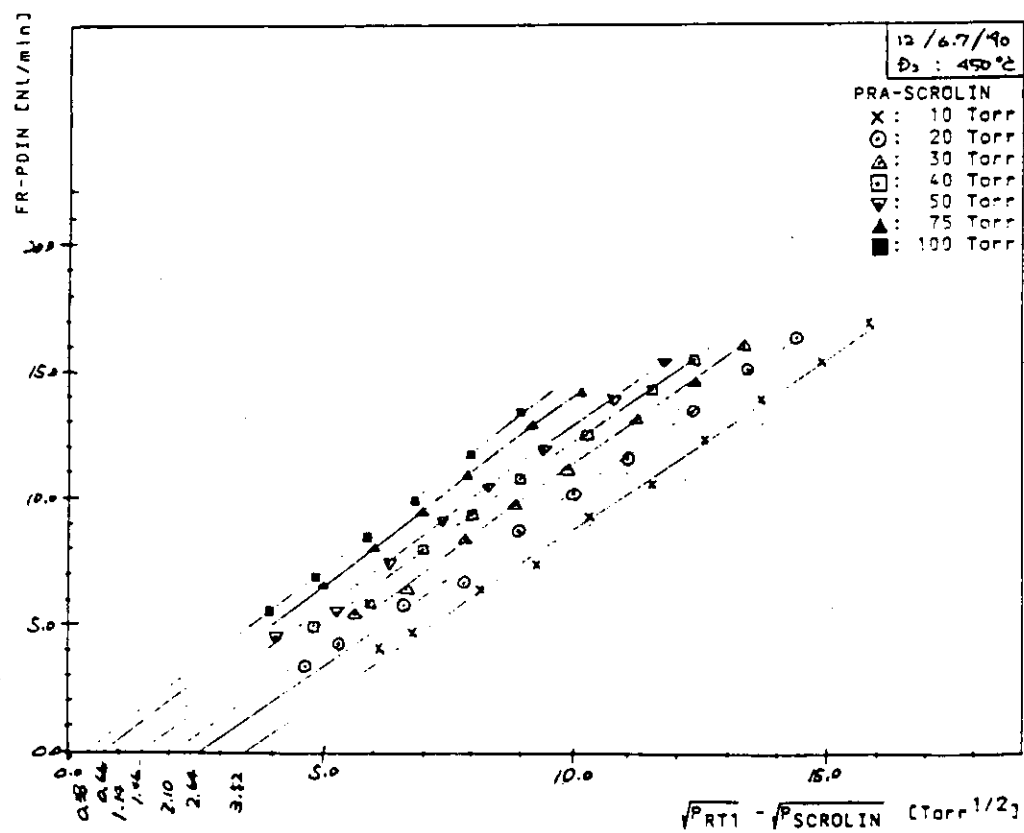
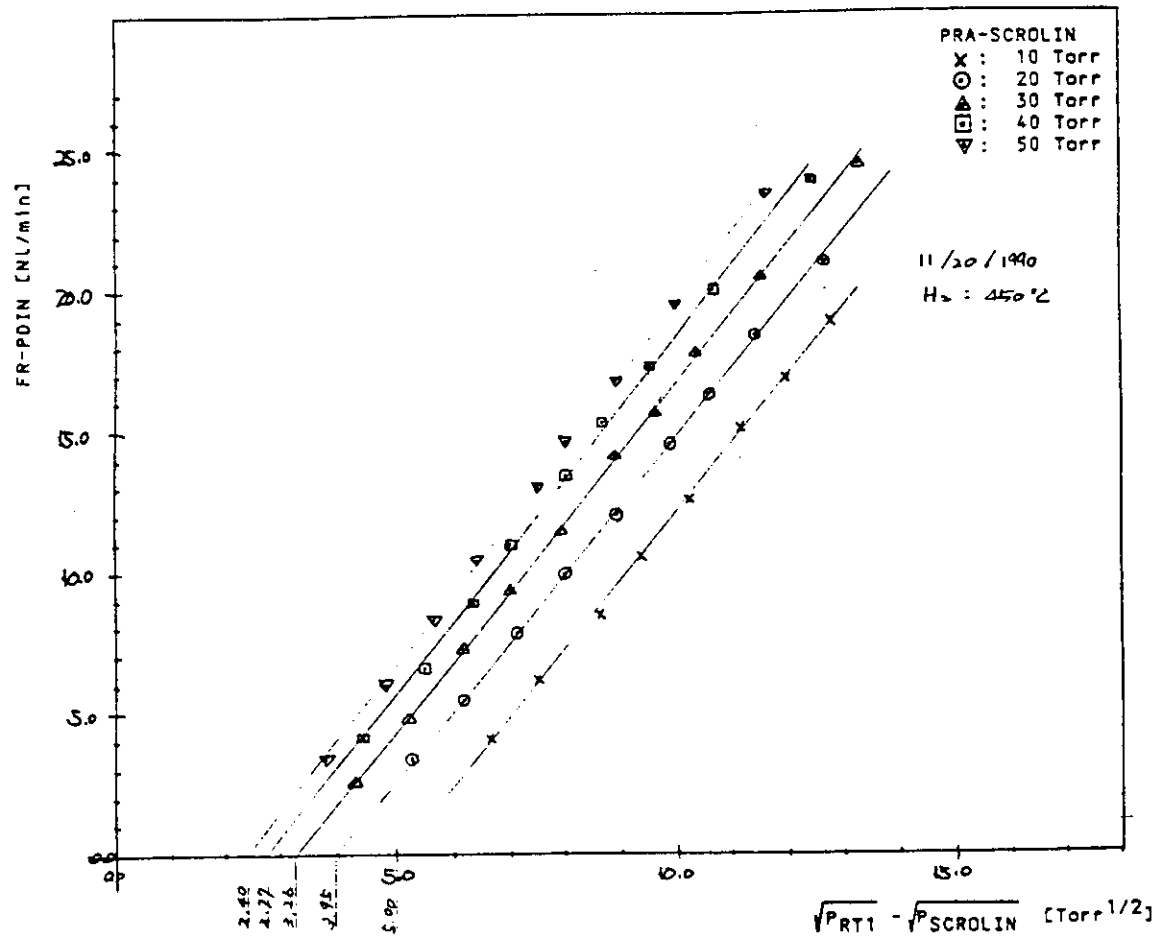


Fig. 2-4 D2,723K

Fig. 2-5 H<sub>2</sub>, 723K (before baking)

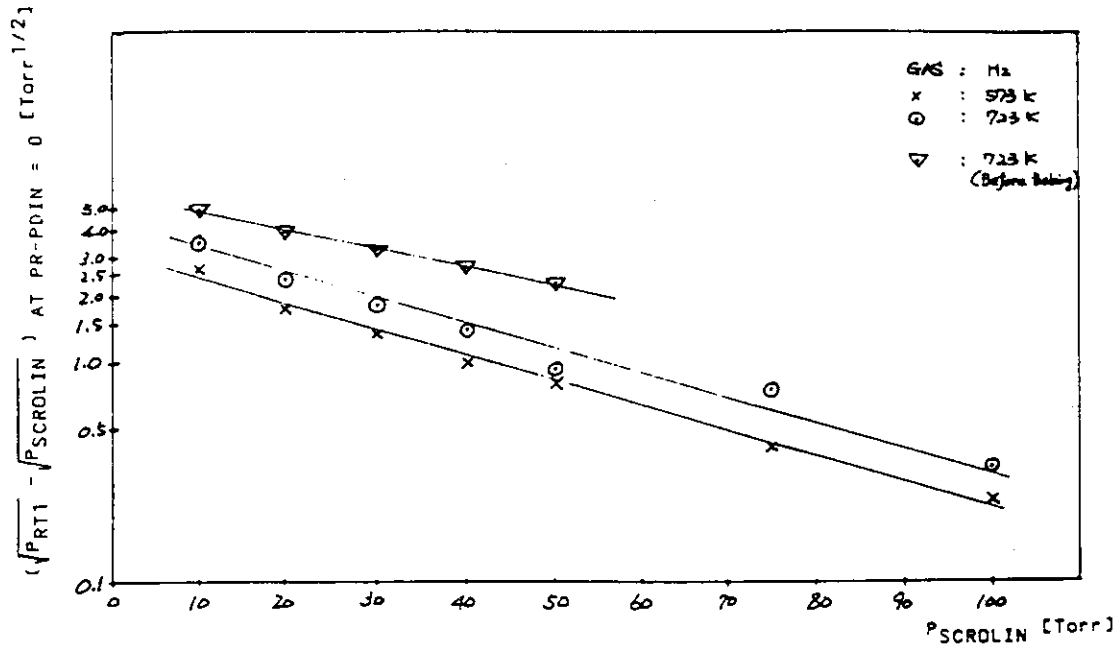


Fig. 3-1 The difference of square root of the pressure at the feed side and permeated side of membrane when the permeation flow rate is zero against the pressure at the permeated side (H<sub>2</sub>)

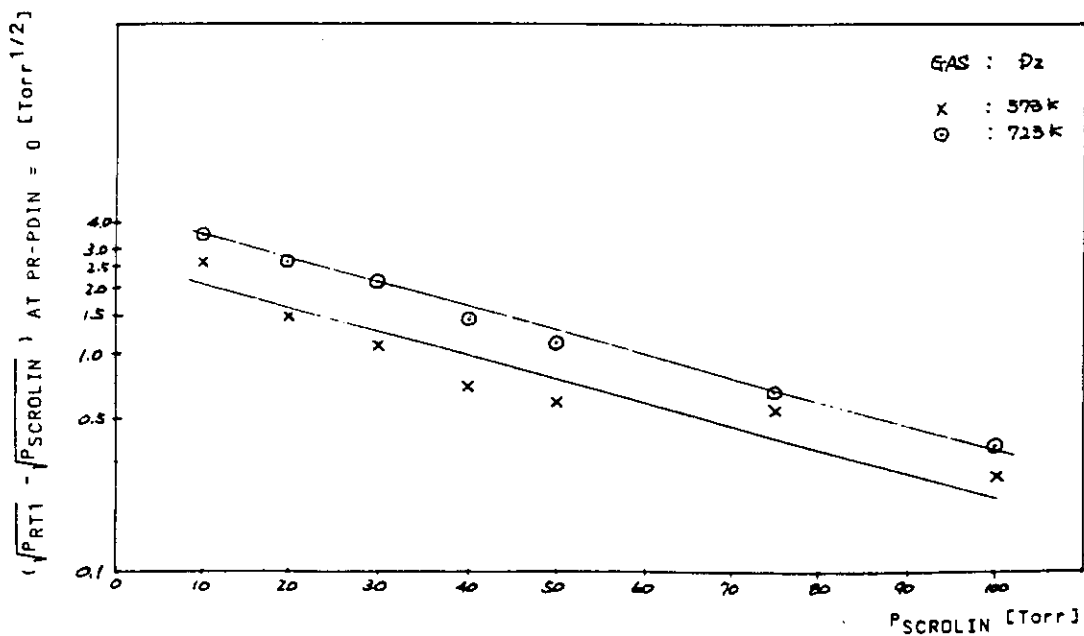


Fig. 3-2 D<sub>2</sub>

#### IV. Conclusion

Pure hydrogen isotopes are circulated through the palladium diffuser and the permeated side was evacuated with the scroll pump. The permeation flux showed linear relation with the differential square-root of pressure across the membrane. Poor permeability was observed especially when the pressure at the permeated side of the membrane is low. It was suspected that a contamination of the surface of the membrane slows down the desorption of hydrogen from the surface and slows reduces permeability. Baking of the membrane at 450°C in oxygen followed by hydrogen reduction improved the permeability. Observed permeability indicates that satisfactory processing rate with tritium can be expected, according to the isotopic ratio of permeability obtained in the previous studies. The linear relationships in the figure showed offsets as a function of the pressure at the permeated side. It is suggested that the reduction of concentration of hydrogen in bleed, that is controlled by the permeation in low (partial) pressure region, may not proceed as expected in high pressure region. Further experiments in conjunction with the performance of the scroll pump and with pure tritium are planned.

## Appendix Consideration on the PD test result

(Appendix to the Watanabe's PD test result report)

S. Konishi

Followings are the discussions and conclusions derived from the test result of the palladium diffuser.

The diffuser showed much more purification capacity than designed for pure  $H_2$  and  $D_2$ . Although permeability of DT in the mixture is yet to be known, the result suggests that the diffuser seems to have capacity as expected.

Permeability of hydrogen isotopes through the membrane increases with temperature, however the change is small. This is well understood that the diffusivity increases with temperature while solubility decreases, thus the permeability as product of these two variables does not change largely with temperature. It will be useful to increase operation temperature when more purification capacity is needed, but will not be very effective.

Linear functions between the permeation flux and the differential square root pressure across the membrane were obtained for each data set with fixed pressure at the permeated side in the pressure region applied in the experiment, that is, under the operation condition of the diffuser. This is mostly explained by the "Sievert's Law", where permeation flux is proportional to the differential square root pressure. This theory is applicable when the diffusion of hydrogen in the metal is the rate controlling process and adsorption/desorption on the metal surface is relatively fast.

Some deviation from the Sievert's Law was observed as the off-set on the extrapolated linear relations crossing zero-flow rate (X-axis). This off-set is nearly zero when the pressure at the permeated side is 100 torr (or higher), and increases as the pressure at the permeated side decreases. It suggests that some mechanisms that disturb desorption of hydrogen from the permeated side of the membrane work (or become the rate controlling step) when hydrogen pressure on it is low.

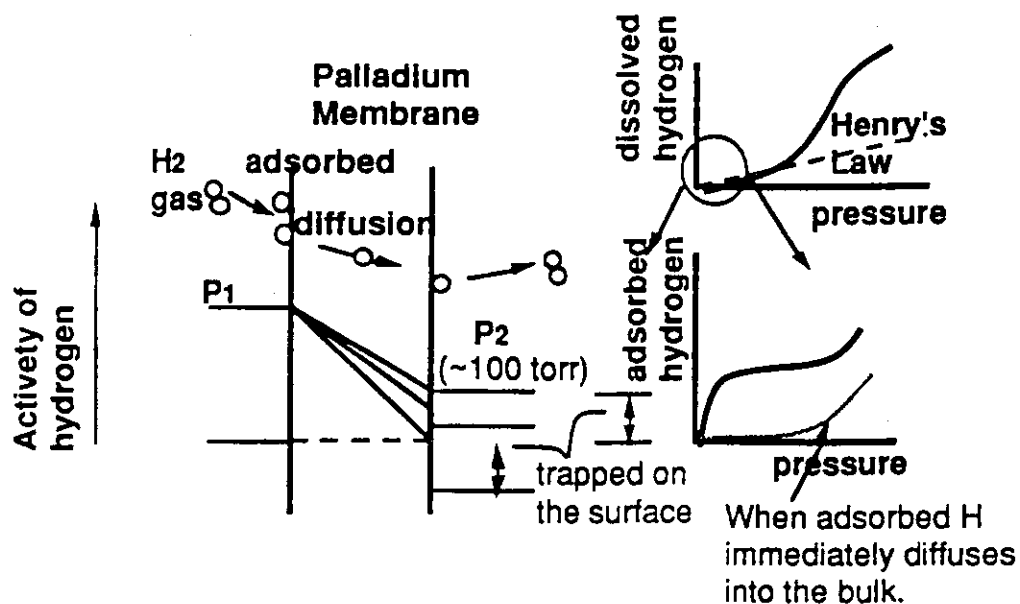
One of the possible explanation for this effect is an existence of hydrogen "Barrier" on the surface of the permeated side of the membrane. This is a layer of adsorbed hydrogen trapped on the surface and does not diffuse into the bulk or recombine to desorb. This mechanism is explained qualitatively in the attached figure.

Baking of the membrane with oxygen at 450°C was found to have an effect to reduce this offset, while hydrogen permeation did not. Some published studies suggest that adsorbed carbon on the permeated surface works as permeation barrier and cannot be removed by hydrogen permeation. The "barrier", probably due to carbon impurity on the membrane, was partially removed by oxygen baking followed by hydrogen reduction, but was still observed in this test after the cleaning.

This result indicates that the permeation does not proceed as fast as in the high pressure region if the hydrogen partial pressures in the feed side and permeated side is lower. In the operation of the diffuser, it means that hydrogen partial pressure in the bleed does not approach zero as expected regardless how low the pressure at the permeated side is. Fortunately, the results of the GC analysis of the bleed gas in the previous cold testings showed a few percent of hydrogen.

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Further implication of the result obtained above is for those who are interested in the use of the diffuser for the "final" tritium recovery. German design of the FCU that utilizes the diffuser combined with catalytic shift reactor expects to extract tritium through the membrane completely before discharging the residues as exhausts. It may take longer to reduce this tritium concentration if it is operated in a batch mode, and certainly it is not suitable for continuous processing.



**Schematic of the hydrogen permeation through palladium and the effect of surface trapping.**

Hydrogen molecules dissociate on the surface of the palladium and then diffuse according to the concentration gradient. Gaseous hydrogen is equilibrated with the hydrogen in/on the metal at the both side of the membrane. Solution of hydrogen into metal usually obeys Henry's Law (dissociation should be accounted), but surface contamination may make hydrogen desorption at the lower pressure side more difficult and/or slower.